

IL NUOVO CIMENTO

ORGANO DELLA SOCIETÀ ITALIANA DI FISICA

SOTTO GLI AUSPICI DEL CONSIGLIO NAZIONALE DELLE RICERCHE

VOL. XVII, N. 5

Serie decima

1° Settembre 1960

On an Attempt of Detection of Primary Cosmic Photons of Very High Energy.

R. MAZE

Cosmic Ray Laboratory, Ecole Normale Supérieure - Paris

A. ZAWADZKI

*Nuclear Research Institute of the Polish Academy of Sciences
Cosmic Ray Laboratory - Lodz*

(ricevuto il 29 Febbraio 1960)

Summary. — In relation to the primary proton flux of very high energy ($(10^{15} \div 10^{16})$ eV), the possible photon flux of equal energy created by the p-p collisions of protons with the diffused matter of the Universe, is evaluated taking into account the multiple production of the π^0 mesons and their decay. If a trace of very high energy photons exists among the primary flux, two types of EAS must be produced in the air: 1) ordinary EAS; 2) pure photon-electron cascades. Taking advantage of the difference between the number of electrons produced at sea level by a protonic and by a photonic EAS of equal energy (therefore resulting in an amplification factor of the observable effect by at least ten times) it is shown that it is possible to separate these two types by studying the penetrating component with the help of a large detection area and to disclose a real limit rate as small as 10^{-4} . Considering current cosmological estimates, it gives some possibility of observing a significant rate. This method therefore may give new experimental information on the origin of cosmic rays and on individual interaction at very high energy.

1. — Evaluation of the possible rate of primary photons of very high energy.

The experiments on extensive air showers, EAS, indicate that primary cosmic rays (probably mostly protons) exist with amounts of energy reaching 10^{19} eV.

It is believed ^(1,4) that such particles possessing energy greater than $(10^{16} \div 10^{17})$ eV do not have a galactic origin as there no sources seem to exist with a vaste and intense enough magnetic field to retain them.

Therefore one can admit that this particle flux is of an extra-galactic origin and that it is the same throughout the Universe.

During their lives the protons of $(10^{17} \div 10^{18})$ eV for example can collide with the diffused matter producing mesons, nucleons and photons (which come from the π^0 -decay) while only the stable particles subsist (protons and photons). Electrons rapidly loose their energy mostly by radiation (magnetic bremsstrahlung) and are considered negligible.

On the other hand, the probability of the destruction of photons by the creation of pairs in hydrogen is about $x/75$, x being the thickness of matter in g cm^{-2} .

The photons have a lower energy than the parents $((10^{15} \div 10^{16})$ eV) as only a small fraction of the interactive energy is transferred to them, and for the observer they are diluted amidst the protons of equal energy.

However, we have undertaken an experiment which should enable us to disclose the existence of such photons if the natural circumstances lend themselves. The chance of success depends upon the value of the product of three evaluations which are only orders of magnitude at the current state of our knowledge; the three factors are the following:

i) *p-p collision probability.* This probability P depends upon the evaluation of the possible crossing of the diffused hydrogen of space by cosmic protons. If the average age of the proton flux is $4 \cdot 10^9$ years and if the average density of matter in the Universe is $\rho = 10^{-27} \text{ g cm}^{-3}$, the protons pass through 4 g cm^{-2} ; according to the mean free path the probability for a collision (p-p) is then 0.1.

From the discovery made at Mt. Palomar of bridges of matter between galaxies and of huge clouds, it is possible that the average density of matter is even bigger than $10^{-27} \text{ g cm}^{-3}$ and more particularly as proposed by ZWICKY ⁽⁵⁾, that $10^{-27} < \rho < 10^{-26} \text{ g cm}^{-3}$ for diffused matter.

ii) *The photon rate in the proton flux.* The evaluation of this rate R_s takes into account:

(1) G. COCCONI: *Suppl. Nuovo Cimento*, **8**, 472 (1958).

(2) K. SITTE: *Suppl. Nuovo Cimento*, **8**, 478 (1958).

(3) F. J. M. FARLEY: *Suppl. Nuovo Cimento*, **8**, 466 (1958).

(4) J. HEIDMAN: *Suppl. Nuovo Cimento*, **8**, 486 (1958).

(5) E. SCHATZMAN: *Origin and Evolution of the Worlds* (Paris, 1957), p. 260.

a) the energy spectrum of the primary particles supposed to be $E^{-1.8}$ for $E > 10^{16}$ eV;

b) the law of multiplicity of production of mesons supposed to vary as $E^{\frac{1}{2}}$;

c) a transfer coefficient (inelasticity parameter) considered as 0.4; those assumptions permit an estimate of the relation between the number of photons and the number of protons of equal energy ($E \geq 10^{15}$ eV) for a collision probability equal to the unity.

This relation between differential spectrums ranges about 10^{-3} . (Calculation in Appendix I).

The rate could be raised by the diffusion effect on protons by extra-galactic fields in an expanding Universe; as suggested by COCCONI⁽¹⁾, when the diffusion speed approaches the recession speed, the contribution to the observed flux of escaping sources is suppressed further away, this means that the secondary photon flux reaches us from a greater space than for protons.

iii) *Factor of displaced energy of photo-electronic EAS in relation to nucleonic EAS.* The detection of very high energy photons can only be done by recording the EAS they produce. A protonic EAS is the result of multiple nuclear interactions and if $E \approx 10^{16}$ eV it has an average density of mesons and nucleons in the vicinity of the core of about several unities to about several tenth of particles per m^{-2} , whereas a photo-electronic cascade shows a density of mesons probably one hundred times smaller (as a consequence of the weak photo-nuclear interaction cross-section),(*) the penetrating particles are therefore practically unobservable in such EAS.

Just for that reason it is possible, thanks to their different morphology, to separate the EAS issued from protons and those issued from photons.

On the other hand the evaluations of the number of electrons at sea level indicate that a purely photo-electronic EAS having the same number of electrons as the protonic EAS, comes from a primary photon with somewhat

(*) The photo-nuclear interaction cross section with air nuclei is about 500 times smaller than that of a proton. A photonic EAS with $N=10^6$ particles at sea level contains $5 \cdot 10^4$ photons of $E \geq 1$ GeV. By considering (however it would be excessive) that this number of photons is conserved for 20 radiation units or 10 nuclear mean free paths, one obtains: $(5 \cdot 10^4 / 5 \cdot 10^2) \cdot 10 = 1000$ interactions with energy greater than 1 GeV. On the other hand a nucleonic shower will contain $8 \cdot 10^4$ mesons, 70% of which have more than 0.8 GeV.

From this we conclude that the photonic shower contains a trifling rate of penetrating particles.

lower energy than that of the primary proton. If this relation of energy is equal to δ , the observation probability of a photonic EAS grows as $a = \delta^\gamma$ times ($\gamma = 1.8$ exponent of the primary spectrum). The factor a (discussed in the Appendix II) probably situated between 10 and 50, facilitates considerably the detection of a photonic EAS and the product of the reckoned values in i), ii), iii), gives:

$$10^{-3} < P \cdot R_s \cdot a < 10^{-2} \quad \text{for } \varrho = 10^{-27} \text{ g cm}^{-3}.$$

Perhaps it is of interest to mention the possibility (as small as it might be) of the existence of galactic photons; obviously, this alternative supposes the necessity of retaining in our galaxy particles of extreme high energy.

Taking into consideration the existence of the galactic halo, one can explain only the presence of nuclei (the magnetic rigidity of which is weaker than that of protons); in this way one is obliged to admit the hypothesis of nuclei as producers of EAS of $E > 10^{16}$ eV.

Assuming a complete fragmentation of a heavy nucleus in the collision with hydrogen, and as a result of this the same photon production as in the single proton nucleon reaction, we obtain the ratio of photons over protons both given by those special events by changing in the above evaluation P by $P \cdot A^{\frac{1}{3}}$ and R_s by R_s/A .

A more complex reaction with nuclear matter and a real fragmentation process would probably increase slightly the $A^{\frac{1}{3}}$ factor, therefore we conclude that the influence of nuclei must be small if there is one.

Whether it involves nuclei or protons, the value of the above mentioned product $P \cdot R_s \cdot a$, if one admits a leakage of photons into outer space which no compensation, will be considerably reduced in the relation $l_\gamma/\lambda_{\text{pp}}$ where l_γ is the galactic mean life of photons ($l_\gamma = 5 \cdot 10^4$ l.y.) and λ_{pp} is the mean free path for the pp collision ($\lambda_{\text{pp}} = 40 \text{ g cm}^{-2}$ or $2.5 \cdot 10^7$ l.y. if $\varrho = 1.6 \cdot 10^{-24} \text{ g cm}^{-3}$ $l_\gamma/\lambda_{\text{pp}}$ is therefore $2 \cdot 10^{-3}$).

If one wishes to obtain a minimum detectable value of an observable rate such as 10^{-3} , more favorable and less probable conditions are then needed for $P \cdot R_s \cdot a$; if in spite of this the experiment shows a sidereal effect on photonic EAS, it will be a proof of galactic origin, as those observed on 50° latitude may come from a distance varying with sidereal time between perhaps $5 \cdot 10^3$ l.y. and $3 \cdot 10^4$ l.y. (direct observation of galactic thickness).

2. - Detecting method for photon-electron EAS.

The experimental setting is composed of two kinds of detectors, one for the registration of the electronic density and consisting in eight banks of nine

counters all unshielded (72 counters of 0.0125 m^2), the other (280 counters of 0.0300 m^2) for the registration of the penetrating component with an effective surface of 7.6 m^2 shielded by a thickness of $25 \text{ cm Pb} + 11 \text{ cm Fe}$.

The probability of detection for a γ -ray in the Compton's window is reduced by 25 cm Pb (counter efficiency supposed to be 0.02) to $1.5 \cdot 10^{-4}$ for 100 MeV and to $2.5 \cdot 10^{-4}$ for 1 GeV . The 11 cm iron sheet under lead reduces this value by more than 10 times; the total reduction is then about $2 \cdot 10^{-5}$ giving for all the surface and for a density of 250 m^{-2} a probability of $4 \cdot 10^{-2}$ for one counter to be hit.

The counters under lead are divided into 56 parts of 5 counters each and the device is driven by a master pulse supplied by an eight-fold coincidence between the eight unshielded banks (when at last one counter is hit in each bank). For each shower the whole registration is made by a hodoscope device, where 128 neon lamps are to be photographed: 72 for the unshielded counters and 56 for the groups of shielded ones.

Thanks to this high degree of surface analysis it is possible to determine exactly the mean density of the electronic and penetrating component. For instance 50 hit unshielded counters give a mean density of about $100 \text{ particles m}^{-2}$, while the probability of registration of a half density by the same counter number diminishes about 10^4 times. In this way the fluctuations of the mean penetrating density Δ_p related to the mean electronic density Δ_e are greatly reduced and a good verification of the Δ_p variation with Δ_e is possible.

To be sure that fluctuations of the number of stricken counters corresponding to a mean give density Δ_p cannot give spurious «zeros» one must choose a minimum electronic density Δ_e and all registrations related to higher values of Δ_e must give then an answer like «yes» or «no» as to the presence of penetrating particles.

The frequency of penetrating events associated to electrons in function of the multiplicity under lead will give a histogram in the form of steps with a maximum and a decreasing frequency on the left going to a low rate corresponding to «3», «2», «1» multiplicity or «zero». Finally, the possible remnant of photonic EAS will be detached by a bigger amplitude for «zero» events as compared to «one» or «two» events; it is in fact an attempt in order to separate a residue by an anticoincidence method as perfect as possible.

In the electronic device, the only spurious events which cannot be eliminated, are due to spurious coincidences between impulses of many counters of individual cells and master pulse, the rate of which is $2.5 \cdot 10^{-2}$ per master pulse, will simulate a hit, so a small amount of real «zeros» will be lost which is without any importance.

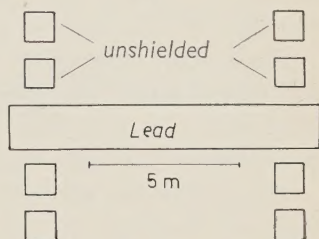


Fig. 1.

The counters which are hit cause the extinction of corresponding neon lamps while the others are lighted; this eliminates completely the possibility of an addition of two successive «zero» events with low electronic density during the time of photographic registration; for other points an efficiency of $(100 \div 10^{-3})\%$ may be proved.

As a short conclusion we hope to have shown that the method may give new experimental information on the origin of cosmic rays and on individual interactions at very high energy, both very difficult problems, and merit to be attempted in spite of an uncertain chance of success.

The device was set up in Lodz at the Cosmic Ray Laboratory of the Nuclear Research Institute of the Polish Academy of Sciences with the help of the Lodz University.

* * *

The authors wish to express particularly their acknowledgements to Professor M. MIĘSOWICZ for stimulating discussions and for his interest in the experiment realized thanks to his help.

APPENDIX I

The photon rate.

In consideration of the variation of multiplicity, the secondary photon spectrum must be slightly steeper than the gerating proton spectrum. Taking

$$(1) \quad N_p dE_p = A E_p^{-(\gamma+1)} dE_p$$

as differential energy spectrum for primary particles, if the multiplicity law for all secondary particles is $m_\pi = K(\eta E_p)^\alpha$, with η inelasticity parameter then the number of secondary particles whose energy is between E_s and $E_s + dE_s$ in

$$(2) \quad N_s dE_s = A K \eta^\alpha E_p^{\alpha-\gamma-1} dE_p.$$

Supposing with a sufficient approximation in our case a symmetric emission and admitting that the energy available in the CM system is equally distributed among the secondaries, remembering that particles are ultra-relativistic, we get for a π -meson

$$(3) \quad E_\pi = \frac{E_p}{E_p^*} E_\pi^* (1 \pm \cos \theta^*),$$

(the asterisk refers to CM system of reference) θ^* is the angle of emission in respect to the incident proton.

Neglecting backward mesons in the CM system and fluctuations, the mean energy of forward mesons which keep the greater part of the energy in the L system becomes:

$$(4) \quad \overline{E}_\pi = \frac{\eta}{m_\pi} E_p (1 + \cos \overline{\theta}^*) .$$

For a very anisotropic emission we take $(1 + \cos \overline{\theta}^*) + C \simeq 1.8$ which gives eliminating m_π with (2) (and leaving out the dash):

$$(4b) \quad E_\pi = \frac{C}{K} \eta^{1-\alpha} E_p^{1-\alpha} = 2E_\gamma$$

the number of photons between E_γ and $E_\gamma + dE_\gamma$ is then

$$(5) \quad N_\gamma dE_\gamma = \frac{1}{3} A K \eta^\alpha E_p^{\alpha-\gamma-1} dE_p .$$

Introducing the variable E_γ given by (4b) in (5) we get

$$(6) \quad N_\gamma dE_\gamma = \frac{1}{3} \frac{A}{1-\alpha} K^{(1-\gamma)/(1-\alpha)} \left(\frac{C}{2} \right)^{(\gamma-\alpha)/(1-\alpha)} \eta^\gamma E_\gamma^{-(\gamma+1-2\alpha)/(1-\alpha)} dE_\gamma ,$$

from which we see that the differential exponent β for the photon spectrum is

$$\beta = \frac{\gamma + 1 - 2\alpha}{1 - \alpha} .$$

Finally we obtain for the spectrum ratio for an equal energy:

$$(7) \quad R_s = \frac{N_\gamma dE_\gamma}{N_p dE_\gamma} = \frac{1}{3(1-\alpha)} K^{(1-\gamma)/(1-\alpha)} \left(\frac{C}{2} \right)^{(\gamma-\alpha)/(1-\alpha)} \eta^\gamma E_\gamma^{-\alpha(\gamma-1)/(1-\alpha)} .$$

Taking $\gamma = 1.8$; $\alpha = 0.25$ and $\eta = 0.5$ with ZATSEPIN⁽⁶⁾ and $K = 1$, one obtains $R_s = 2.2 \cdot 10^{-3}$ for $E_\gamma = 10^6$ GeV and $R_s = 1.2 \cdot 10^{-3}$ for $E_\gamma = 10^7$ GeV thus in the energy range which we considered: $R_s \simeq 10^{-3}$.

Notice that we did not take into account the production of K-mesons, baryons and antibaryons which may lower the R value by about 20%; besides the multiplicity may increase less steeper than $E^\frac{1}{2}$ giving a sensitive increase of R_s (for instance if $\alpha = 0.2$ one obtains an increase of nearly three times for $E_\gamma = 10^{16}$ eV); notice also that energy equipartition hypothesis is the most unfavourable because R_s must increase as a more important part of energy is transferred to a smaller number of mesons.

(6) G. T. ZATSEPIN: *International Cosmic Ray Conference* (Moscow, 1959), Abstracts pp. 15-16.

For an example of the application of the formula (7) taking $\eta = 0.3$ and $\gamma = 1.6$ as known by the study of jets in emulsions for secondaries of 10^{12} eV, one gets for photons $R_s(\gamma) = 1.3 \cdot 10^{-2}$ and for charged pions $R_s(\pi_{\pm}) = 5 \cdot 10^{-2}$ [$R_s(\pi^{\pm}) = 2^{(\gamma-\alpha)/(1-\alpha)} R_s(\gamma)$] by introducing the variable E_{π} given by (4b) in (5).

This ratio agrees well with the ratio deduced at this energy from calculations^(7,8) of the generation pion spectrum in the air with the help of the muon spectrum as measured in emulsions exposed at high altitude.

APPENDIX II

On the amplification factor a .

A calculation of the number N of electrons at sea level and for two primary protons of 10^{13} and 10^{15} eV energy, has been presented by UEDA and OGITA⁽⁹⁾ for three values of the inelasticity parameter.

It would be interesting to compare the N values given by the cascade theory used by these authors for a single primary photon of the same energy. To look at this question more precisely we have calculated in both cases the N value for a 10^{16} eV primary with the help of the Snyder theory in B approximation: in the protonic EAS, this theory is applied to photons issued from π^0 created in successive collisions. The difference between our assumptions and those of Ueda-Ogita lies principally on the regeneration of pions by pions supposed to be completely inelastic as indicated by the Bristol Group⁽¹⁰⁾. As for the rest, our calculation is characterized as follows:

- a) the collision mean free path is equal to two radiation units;
- b) the pion decays and photons the energy of which is less than 10^{10} eV give no cascade;
- c) the multiplicity law is $m = (\eta E)^{\frac{1}{2}}$.

For $\eta = 0.5$ one obtains the following results at sea level:

Photonic EAS	Photonic EAS
$1.2 \cdot 10^6$ particles	$4 \cdot 10^6$ particles .

As this scheme gives an attenuation path and a mean age for the electronic component in rather good accordance with experimental data, our feeling is

⁽⁷⁾ G. PUPPI: *Progress in Cosmic Ray Physics*, **3**, 341 (1956).

⁽⁸⁾ A. SUBRAMANIAN and S. D. VERMA: *Nuovo Cimento*, **13**, 572 (1959).

⁽⁹⁾ A. UEDA and N. OGITA: *Progr. Theor. Phys.*, **18**, 269 (1957).

⁽¹⁰⁾ B. EDWARDS, J. LOSTY, D. H. PERKINS, K. PINKAU and J. REYNOLDS: *Phil. Mag.*, **3**, 237 (1958).

that a photonic EAS of 10^{16} eV contains three or four times (*) more electrons than a protonic one. By taking into account the exponent of the primary energy spectrum one obtains then an amplification factor of about 10 ($a = \delta''$).

(*) If $\eta=1$ this value of δ becomes seven in accordance with BUDINI and MOLIÈRE therefore a becomes about thirty (*Kosmische Strahlung*, W. HEISENBERG editor, Berlin, 1953).

Note added in proof.

It is known at present that in spite of the large value of the total inelasticity coefficient which is about 0.4, the π -meson production coefficient should probably not exceed 0.2.

The resulting decrease of the effect described by us will be however partially compensated, as on the other hand it appears now that a single meson may carry sometimes a considerable part of the energy of secondary particles.

RIASSUNTO (*)

In relazione al flusso primario di protoni di altissima energia ($(10^{15} \div 10^{16})$ eV) si valuta il possibile flusso di fotoni di uguale energia creato da collisioni p-p di protoni con la materia diffusa dell'Universo, tenendo conto della produzione multipla dei mesoni π^0 e del loro decadimento. Se nel flusso primario esiste una traccia di fotoni di altissima energia, si possono produrre nell'aria due tipi di EAS: 1) EAS ordinario; 2) pure cascate fotone-elettrone. Avvalendosi della differenza fra i numeri di elettroni prodotti al livello del mare da EAS protonici ed EAS fotonici di uguale energia (dalla quale risulta un fattore di amplificazione degli effetti osservabili di almeno 10) si mostra che è possibile separare questi due tipi osservando la componente penetrante con l'ausilio di una grande area di rivelazione e determinare un rapporto limite reale sino ad un minimo di 10^{-4} . Considerando le correnti valutazioni cosmologiche, il metodo dà qualche possibilità di osservare un rapporto significativo. Questo metodo quindi può dare nuove informazioni sperimentali sull'origine dei raggi cosmici e sulle singole interazioni di altissima energia.

(*) Traduzione a cura della Redazione.

Angular Distributions of Protons Emitted in $^{24}\text{Mg}(\text{n}, \text{p})$ and $^{32}\text{S}(\text{n}, \text{p})$ Reactions at 14 MeV.

L. COLLI

Laboratori CISE - Milano
Istituto di Fisica dell'Università - Milano

I. IORI, G. MARCAZZAN, F. MERZARI, A. M. SONA (*) and P. G. SONA

Laboratori CISE - Milano

(ricevuto il 2 Aprile 1960)

Summary. — In this paper measurements are described on angular distributions of protons from the reactions $^{32}\text{S}(\text{n}, \text{p})$ and $^{24}\text{Mg}(\text{n}, \text{p})$, with 14 MeV energy neutrons. For each element the angular distribution is studied for a well defined group of protons, corresponding to a level group which originate a gross-structure. Experimental results allow the following conclusions: 1) the reaction mechanism for protons in the gross-structures is certainly a direct surface effect; 2) it is possible to determine the l values of orbits from which the proton is taken and where the neutron is captured. In both cases a part of the spectrum can be attributed to a statistical evaporation process.

Recent studies on reactions (n, p) at 14 MeV have established the existence of gross-structures in the spectrum of protons emitted by some nuclei ⁽¹⁾. An interpretation of these gross-structures was given in that paper, and they were attributed to groups of levels of the residual nucleus more favoured in

(*) Now at the University of Florence.

⁽¹⁾ L. COLLI, F. CVELBAR, S. MICHELETTI and M. PIGNANELLI: *Nuovo Cimento*, **14**, 81 (1959).

the (n, p) transition and corresponding to the position of single particle levels of that nucleus.

For the simple fact that some level groups are more favoured than others it should be supposed that the mechanism by which the reaction takes place is one of the direct interaction type, as no similar result can be explained by a mechanism of the evaporative type.

The measurement of the angular distribution of these particularly favoured level groups may enable us to verify the hypotheses advanced so far.

The measurement of the angular distribution gives in fact the following indications:

1) If no isotropy or symmetry with respect to 90° exists, the presence of an interaction mechanism of the direct type is assured.

2) The comparison of the anisotropic part with the theories developed for the direct effects (BUTLER ⁽²⁾, GLENDENNING ⁽³⁾) enables us to establish the value of the orbital angular momentum given to the nucleus in the transition.

The shell model furnishes a description of single particle levels, on which the particles that take part in the reaction can be found.

If the reaction evolves preferentially by giving rise to transitions between the single particle levels occupied by the proton and the neutron, the measurements of the transferred orbital momentum allows to know the single particle states between which the transition took place.

For all these reasons, we have begun a series of measurements of the angular distribution of the protons corresponding to the gross-structure. In this work we are presenting the result of the measurements on the peak of $^{24}\text{Mg}(n, p)$ at 1.8 MeV residual nucleus excitation energy and of $^{32}\text{S}(n, p)$ at 4.2 MeV excitation energy.

1. - Description of apparatus.

The neutrons, of energy 14.1 MeV, are obtained by means of the reaction ($d+t$). The proton detector is the same described in previous works ⁽⁴⁾, consisting of a telescope made of two proportional counters and a scintillation counter in coincidence, plus a proportional counter in anticoincidence.

⁽²⁾ S. T. BUTLER: *Nuclear Stripping Reactions* (New York, 1957).

⁽³⁾ N. K. GLENDENNING: *Phys. Rev.*, **114**, 1297 (1959).

⁽⁴⁾ G. MARCAZZAN, M. PIGNANELLI and A. M. SONA: *Nuovo Cimento*, **10**, 155 (1958).

The pulses given by the photomultiplier are amplified with a normal electronic chain and analysed with a 100-channels pulse analyser.

With such instrument, the energy resolution obtained is between 4 and 5%; all events due to α particles and electrons are excluded. To measure the angular distribution, all the apparatus is turned keeping fixed the center of the target and rotating the telescope in respect of the beam of incident neutrons. The angular aperture with which the protons are detected is 10° .

The energy spectrum of the emitted particles is taken at the choosed angle. All the spectra are given in the energy scale referred to the center of mass system.

It is easy to recognize in the spectra so obtained at various angles the peak corresponding to the gross-structure under study, and by counting the number of events contained in the peak at various angles we can obtain the angular distribution of the corresponding protons.

The calibration of the energy scale is made measuring the height of the pulses given by the recoil protons corresponding to the 14 MeV neutrons obtained from a thin polythene target.

The measurement of the intensity of the beam of incident neutrons is obtained by counting the number of recoil protons given by the same polythene target. It is thus possible to calculate the absolute value of the cross-section of the reaction under study.

2. - Description of results.

$^{32}\text{S}(n, p)^{32}\text{P}$. The experimental results of the measurements on protons from $^{32}\text{S}(n, p)$ reaction at 11 different angles are included in a previous paper ⁽⁵⁾ where we have discussed the angular distribution of deuterons from the $^{32}\text{S}(n, d)$ reaction. In fact, as we did not discriminate particles of different masses, the published spectrum contains both protons and deuterons.

In the course of more detailed investigations ⁽¹⁻⁶⁾ it has been shown that the spectrum contains a proton group of 8.2 MeV energy and a deuteron group of 6.6 MeV energy. We are considering now the proton group.

The proton peak considered corresponds to about 4.2 MeV excitation energy of the residual nucleus, and its width is about 1.5 MeV. From the known spectrum of the excitation levels of the residual nucleus ^{32}P , we know that

⁽⁵⁾ L. COLLI, G. MARCAZZAN, F. MERZARI, P. G. SONA and F. TONOLINI: to be published in *Nuovo Cimento*.

⁽⁶⁾ L. COLLI, F. CVELBAR, S. MICHELETTI and M. PIGNANELLI: *Nuovo Cimento*, **13**, 868 (1959).

this peak can contain the levels at 3.45, 4.03, 4.21, 4.43, 4.9, 5.11, 5.37, 5.53 MeV.

Fig. 1 shows the experimental angular distribution of the proton group, calculated by taking account of the protons of energy contained between 6.8 MeV and 9 MeV.

By observing the angular distribution shown in Fig. 1 we can see:

- 1) that there is a part strongly anisotropic and concentrated at small angles;
- 2) that the angular distribution is practically constant for angles greater than 80° , around the cross-section value of 4 mb/sr.

The errors given with the experimental points are the statistical ones. In comparison with these all the other errors should be considered negligible.

The anisotropy of the angular distribution gives evidence of a direct effect.

In order to explain our results, we have considered the theory of direct effects developed by BUTLER⁽²⁾. According to this theory, the interaction occurs directly with a proton of the nuclear surface, without interaction with the rest of the nucleus.

In the case of an (n, p) reaction, this theory contemplates an angular distribution given simply by:

$$(1) \quad \sigma(\theta) \div \sum_l C_{l_n l_p}^2(l, 0; 0, 0) j_l^2(QR) \left| \left\langle \frac{\mathbf{K}'}{2} \middle| r_{np} \middle| \frac{\mathbf{K}'}{2} - \mathbf{K} \right\rangle \right|^2,$$

where $C_{l_n l_p}(l, 0; 0, 0)$ are Clebsch-Gordan coefficients and j_l the spherical Bessel functions, being l_n and l_p the angular momenta of the neutron and the proton, and l the angular momentum transferred in the reaction. The sum must be

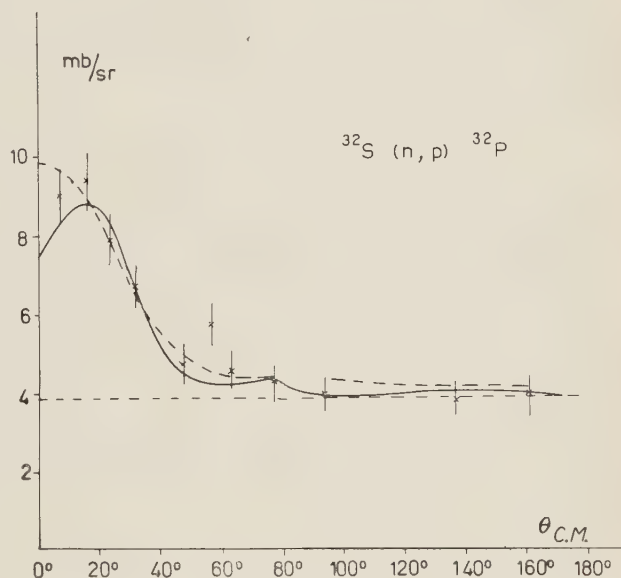


Fig. 1. - Angular distribution of the proton group at 8.2 MeV from $^{32}\text{S}(n, p)^{32}\text{P}$ reaction. The dotted line is calculated following Butler's theory taking for transferred l values 0, 2, 4, $R=3.8 \cdot 10^{-13}$ cm and arbitrarily normalized. The full line is calculated following Butler's theory with $l=1$, $R=7 \cdot 10^{-13}$ cm and arbitrarily normalized. Smearing for angular aperture is included.

extended to all l -values allowed by the angular momentum conservation laws.

R is the radius of interaction and $Q = |\mathbf{K} - \mathbf{K}'|$ where \mathbf{K} and \mathbf{K}' are the wave vectors for the incident and outgoing particles respectively; the factor $\langle \mathbf{K}'/2 | r_{np} | (\mathbf{K}'/2) - \mathbf{K} \rangle$ is the matrix element of the r -matrix which can be determined phenomenologically from two body scattering measurements; in fact this factor is obtained from the neutron-proton scattering cross-section through the relation

$$\sigma_{np} = \frac{2\pi}{\hbar^2} m^2 \left| \left\langle \frac{\mathbf{K}'}{2} \left| r_{np} \right| \frac{\mathbf{K}'}{2} - \mathbf{K} \right\rangle \right|^2.$$

Considering the experimental points with their errors at small angles, we cannot establish if the maximum of the angular distribution curve is at 0° or at 15° .

The experimental points can be fitted either taking an $l_{\min} = 0$ transition with an R value $3.8 \cdot 10^{-13}$ cm or an $l = 1$ transition with an R value $7 \cdot 10^{-13}$ cm. Fig. 1 shows the two curves together with the experimental points. The curve corresponding to $l_{\min} = 0$ contains the contribution of the j_2^2 and j_4^2 terms as given in Butler's formula, in the case that these l values are allowed by the angular momentum conservation rules. However the contribution of these two terms is quite small.

The value of the σ_{tot} , for the peak studied, as derived from the integration on the angles of the spectra, is (57.7 ± 3) mb, 43.3 of which is due to the possible evaporated part and 14.4 to the direct part.

$^{24}\text{Mg}(n, p)^{24}\text{Na}$. Fig. 2 compiles all the measurements made with ^{24}Mg . 8 angles were studied between 7° and 135° .

The angular distribution was calculated for the peak of protons at 6.8 MeV, which corresponds to 1.8 MeV excitation energy of the residual nucleus. This peak can contain the three known excitation levels of the residual nucleus at 1.34, 1.84, 1.88 MeV.

In view of the high threshold value for the (n, d) reaction, these spectra do not contain deuterons.

The protons peak, is of about 1 MeV width. Fig. 3 reports the angular distribution from the spectra of Fig. 2.

This angular distribution shows an anisotropy with greater probability of emission forward; two steps are clearly visible, one at 45° and the other at 90° .

This angular distribution cannot be explained by means of a simple Butler's curve j_l^2 . The position of the first step corresponds well with the maximum of the j_2^2 curve (that is a transition with $l = 2$) when we take for the interaction radius the value $R = 6.5 \cdot 10^{-13}$ cm, about one unity bigger than the Gamow-Critchfield radius.

The whole curve can be fitted if we take into account a small contribution from the j_0^2 ($l = 0$ transition) and also a j_4^2 contribution ($l = 4$ transition) as

given by the formula:

$$\frac{1}{10}j_0^2 + 1j_2^2 + 2j_4^2.$$

The coefficients are chosen to the best fit.

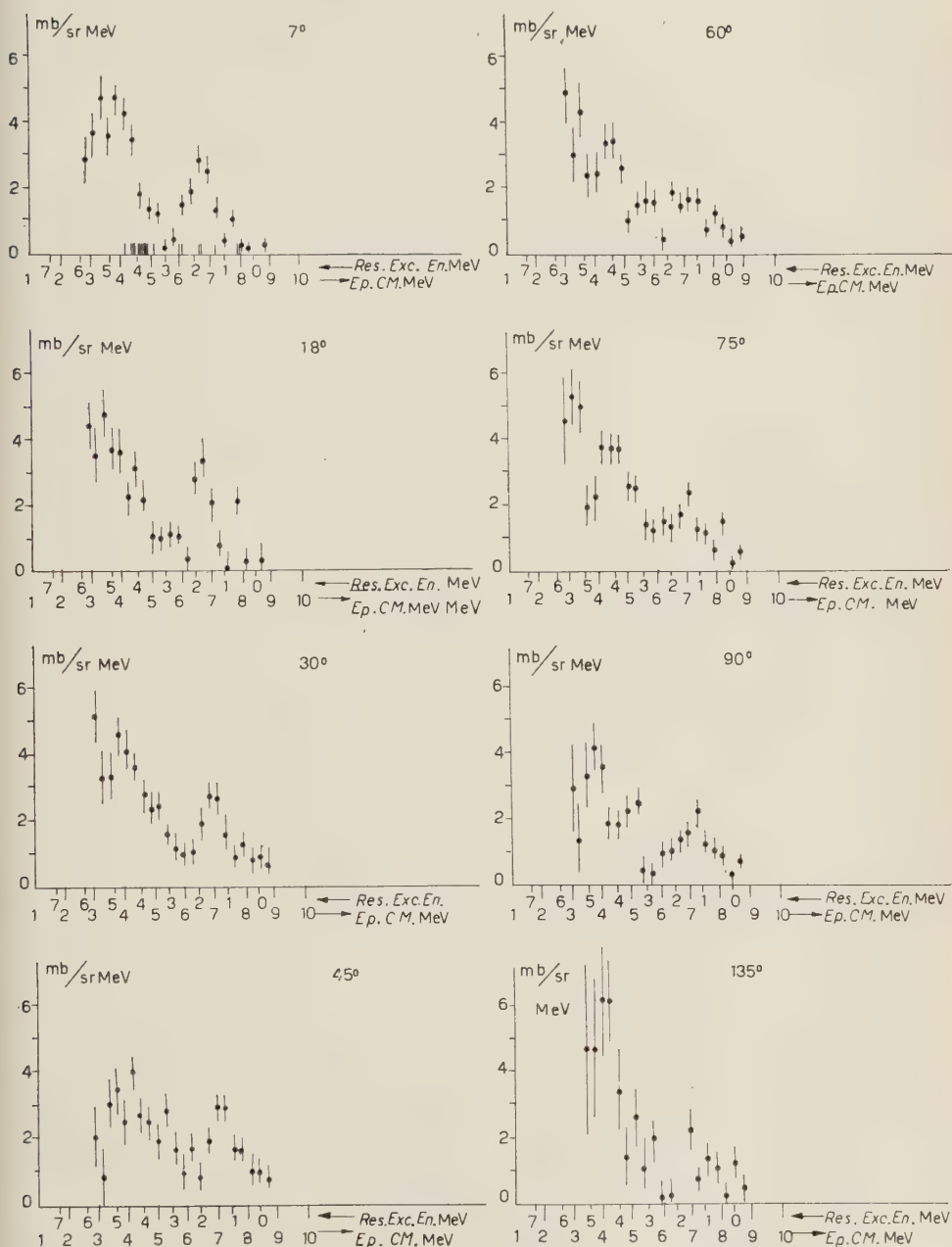


Fig. 2. - Spectra of protons taken at various angles from the reaction $^{24}\text{Mg}(n, p)^{24}\text{Na}$.

The value of the σ_{tot} for the peak of protons under discussion turns out to be (28 ± 2) mb.

Another possibility to be taken into account when interpreting these results

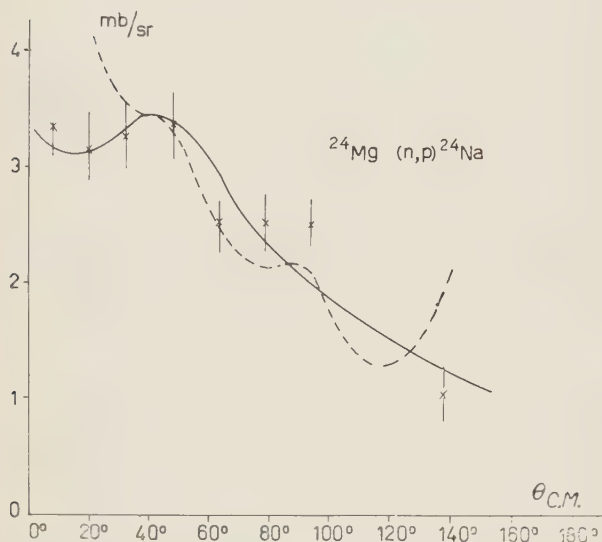


Fig. 3. - Angular distribution of the proton group at 6.8 MeV from the $^{24}\text{Mg}(n, p)$ reaction. The dotted line is the one given by Glendenning in a very similar case, and for a transition with $l=2$, arbitrarily normalized. The full line is calculated with the formula $(\frac{1}{10}J_0^2(QR) + J_2^2(QR) + 2J_4^2(QR))$ with $R = 6.5 \cdot 10^{-13}$ cm and arbitrarily normalized.

distribution quite similar to that of Fig. 3, in the case where the l value is 2.

In this order of ideas, we could attribute to the transition the value of $l=2$.

However the correct application of Glendenning's theory to the case of $^{24}\text{Mg}(n, p)$ requires a long series of calculations, now in progress.

3. - Discussion

3.1. *Direct effect.* - In the case of sulphur, we observe an angular distribution which can be interpreted by means of Butler's formula either with $l_{min}=0$, or with $l=1$. It is therefore quite probable that the mechanism of

is to consider the distortion effect on the incident and outgoing waves due to the optical potential of the nucleus. It is well known in fact that the wave distortion causes a forward concentration in particle emission, even in the case where the transferred angular momentum is $l > 0$. This result was found by LEVINSON and BANERJEE⁽⁷⁾, as well as by GLENDENNING⁽³⁾. If considering the case of $\text{Mg}(n, p)$ we can observe that the calculations made by GLENDENNING under the hypothesis of surface interaction for inelastic scattering of 15 MeV neutrons on a nucleus with $A=30$, contemplate an angular

⁽⁷⁾ C. A. LEVINSON and M. K. BANERJEE: *Am. Phys.*, **2**, 471 (1957); **2**, 499 (1957); **3**, 67 (1958).

the reaction for the anisotropic part is a direct interaction, which induces transitions between the single particle states of the neutron and the proton of the nucleus. In what follows we shall consider separately the two possible l values.

1) $l = 0$. On the basis of the shell model, it is found that the only possibility of a transition with $l_{\min} = 0$ taking place by leaving the residual nucleus in an excited state of a few MeV, that is between not too distant orbits, can occur between a proton occupying a $d_{3/2}$ orbit and a neutron occupying a $d_{3/2}$ orbit.

In fact, the nucleus ^{32}S has its last two protons and two neutrons on the shell $s_{1/2}$ which is therefore complete; the first empty orbit for neutrons (or protons), is the $d_{3/2}$, and the penultimate full orbit for protons (or neutrons) is $d_{3/2}$.

The transition which leads to the fundamental states of the residual nucleus ^{32}P can take place when a proton is made to be emitted from the orbit $s_{1/2}$ and a neutron captured on the orbit $d_{3/2}$. We can obtain an excited state by letting a proton be emitted by the orbit $d_{3/2}$ and a neutron captured in the orbit $d_{3/2}$. In this transition $l_{\min} = 0$. The residual nucleus ^{32}P in this case has an empty place for protons in the orbit $d_{3/2}$ and two protons in the $s_{1/2}$ orbit. The angular momentum conservation allows l -values 0, 2, 4 as taken into account calculating the curve for $l_{\min} = 0$ of Fig. 1 following Butler's formula (1).

2) $l = 1$. In this case we can obtain such transition taking out from the sulphur nucleus the last $s_{1/2}$ proton and putting a neutron on the $p_{3/2}$ orbit, so leaving two neutron orbits free ($d_{3/2}$ and $f_{7/2}$). This is a single particle excited state, and the only possible l value is $l = 1$.

This last case $l = 1$ seems to us the most probable one; this opinion is supported by the fact that also the analysis of angular distribution in the case of Mg requires a rather big value of the interaction radius. We have further evidence of big interaction radius in the case of Si (n, p). These measurements will be published shortly.

The considerations previously done on $^{24}\text{Mg}(n, p)^{24}\text{Na}$ results show that it is not possible to fit our experimental points with a curve calculated from a simplified theory as Butler's theory is. In fact, either distorted wave effect or contribution from 0 and 4 l -values must be taken into account, and it must be noted that this contribution is in different quantity as calculated from Butler's formula (1), which gives always a much bigger contribution from $l = 0$, compared to what we can put to fit our curve. So, an alternative explanation of this result, beside the distorted wave effect, is to assume a pre-dominant $l = 2$ transition with some $l = 0$ due to some kind of configuration

mixing; this is not improbable, because ^{24}Na is certainly a deformed nucleus; Nilsson's model supports this point of view.

In any way, we can deduce that the $l = 2$ transition is the most important. This corresponds to a reaction taking out a proton from the last shell $d_{3/2}$ and putting a neutron on the shell $s_{1/2}$.

So we have an excited single particle state at 1.8 MeV.

Following the interpretation we have given to our experiments, we can deduce from them the position of some shell model orbits in ^{24}Na and ^{32}P nuclei; namely the distance between $d_{3/2}$ and $s_{1/2}$ neutrons orbits in ^{24}Na is 1.8 MeV and the distance between $d_{3/2}$ and $p_{3/2}$ neutron orbits in ^{32}P is 4.2 MeV.

3'2. Evaporation. — We should like now to discuss the parts of the spectra with isotropic angular distribution.

The measurements made on sulphur show that both the protons emitted by direct effect and the deuterons emitted by pick-up process are very much concentrated forward (⁵).

We may therefore consider the spectra obtained at back angles being practically due to an evaporation of protons. Above 60° , the spectrum has in fact a smooth shape, without peaks. Analysing this spectrum under the evaporation theory, a temperature of 1.45 MeV is found in good agreement with the values obtained for nuclei of neighbouring masses (⁶). The cross-section value of the evaporation process discussed here may be easily obtained from the spectra.

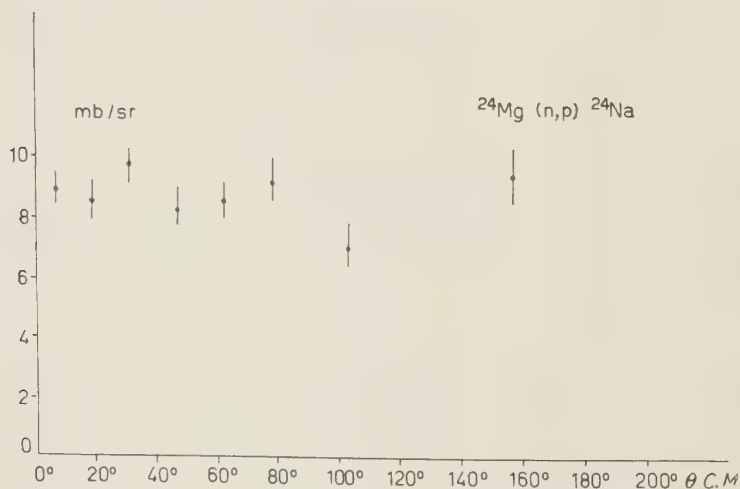


Fig. 4. — Angular distribution of the low energy part of the proton spectrum from $^{24}\text{Mg}(n, p)^{24}\text{Na}$.

(⁶) T. ERICSON: *Nucl. Phys.*, **11**, 481 (1959).

It is calculated a $\sigma_{\text{evap}} = (285 \pm 20)$ mb. The eventual contribution of the reaction (n, np) is negligible, as protons due to this reaction are possible only below 4.5 MeV.

It is interesting to note that the evaporation contributes to a greater extent to the σ_{tot} of the reaction (n, p) than the direct effect which, though considerable, is limited to the forward angles that have little weight on the integration over the angles.

The spectrum of protons emitted by ^{24}Mg features, similarly, an isotropic part, below 6 MeV, which can also be explained with an evaporation process with a $\sigma_{\text{evap}} = (109 \pm 10)$ mb. (See Fig. 4).

By analysing it in this sense, an evaporation temperature of 1.2 MeV is obtained.

In this case the spectra at backward angles do not have a smooth shape, still showing gross-structures, so we cannot be sure that the low energy peak is due to evaporation rather than to some direct effect, whose angular distribution is badly distorted due to Coulomb's interaction or other similar effects.

* * *

We gladly thank Prof. U. FACCHINI for the many discussions throughout the work.

RIASSUNTO

In questo lavoro sono descritte le misure della distribuzione angolare dei protoni emessi nelle reazioni $^{32}\text{S}(\text{n}, \text{p})$ e $^{24}\text{Mg}(\text{n}, \text{p})$. In ognuno dei due casi viene particolarmente studiata la distribuzione angolare di un ben definito gruppo di protoni, corrispondenti ad un gruppo di livelli che dà origine ad una «gross-structure». I risultati sperimentali portano alle seguenti conclusioni: 1) il meccanismo della reazione per i protoni compresi nei gruppi delle «gross-structures» è certamente un effetto diretto, di superficie; 2) è possibile determinare le orbite da cui viene emesso il protone e in cui viene catturato il neutrone. In ambedue i casi una parte dello spettro potrebbe essere dovuta ad un processo di evaporazione statistica.

On the Non-Orthogonality Problem in the Semi-Empirical MO-LCAO Method.

G. DEL RE (*)

Quantum Chemistry Group, University of Uppsala - Uppsala

(ricevuto l'8 Aprile 1960)

Summary. — An explicit correlation between semi-empirical MO-LCAO calculations for π -systems including and not including overlap is derived and discussed under the assumption that overlap integrals, when taken into account, are proportional to the corresponding bond integrals. It is shown that, so far as atomic charges are concerned, the introduction of overlap makes practically no difference in the results. On the contrary all other quantities undergo sometimes serious changes. This is shown by means of a few numerical examples; it gives some support to the idea that the practice of neglecting overlap may not be so sound as has been believed.

1. — Introduction.

As is well known, in the MO-LCAO method, a molecular orbital Ψ_i is expanded in a basis Φ formed by a set of «atomic Orbitals» (AO) Φ_1, Φ_2, \dots , i.e. of (normalized) functions centered on the various atoms of the molecule under study, and sharing at least the symmetry properties of one-centre eigenfunctions:

$$(1) \quad \Psi_i = \sum_{\mu} \Phi_{\mu} c_{\mu i}.$$

By use of matrix formalism and of the variation principle, the evaluation of

(*) Now at RIAS, Baltimore, Md., USA; on leave from Scuola di Perfezionamento in Fisica Teorica e Nucleare, University of Naples, Naples, Italy.

the coefficients $c_{\mu i}$ is reduced to the problem of solving the equation

$$(2) \quad \mathbf{H}\mathbf{C} = (1 + \mathbf{S})\mathbf{C}\epsilon,$$

where \mathbf{C} is a matrix whose elements are the $c_{\mu i}$'s, so that:

$$(3) \quad \Psi = (\Psi_1, \Psi_2, \Psi_3, \dots) = (\Phi_1, \Phi_2, \dots)\mathbf{C} = \Phi\mathbf{C}.$$

\mathbf{H} and \mathbf{S} are defined, in terms of the effective Hamiltonian H_{eff} and of the atomic orbitals, as

$$(4) \quad \begin{cases} (\mathbf{H})_{\mu\mu} = \alpha_{\mu} = \int \Phi_{\mu}^* H_{\text{eff}} \Phi_{\mu} d\tau & \text{(Coulomb integral),} \\ (\mathbf{H})_{\mu\nu} = \beta_{\mu\nu} = \int \Phi_{\mu}^* H_{\text{eff}} \Phi_{\nu} d\tau & \text{(bond-integral);} \end{cases}$$

$$(5) \quad \begin{cases} (\mathbf{S})_{\mu\mu} = 0, \\ (\mathbf{S})_{\mu\nu} = \int \Phi_{\mu}^* \Phi_{\nu} d\tau & \text{(overlap integral);} \end{cases}$$

ϵ is the diagonal matrix whose non-zero elements are the orbital energies $\epsilon_1, \epsilon_2, \dots$.

The problem of solving approximately a molecular problem is thus reduced to the problem of evaluating the elements of \mathbf{H} and of \mathbf{S} , and of solving the systems of equations in the $c_{\mu i}$'s to which (2) is equivalent.

In the present paper, we shall be concerned only with one of these problems, that connected with the presence of \mathbf{S} in equation (2); therefore, we are not directly concerned with the problem of evaluating \mathbf{H} . However, since our treatment implies the use of an empirical assumption about the connection between \mathbf{S} and \mathbf{H} , we shall discuss our procedure in strict connection with the semiempirical MO-LCAO method. The latter, as is well known, has been remarkably successful in applications, and therefore it seems to us that it is worthwhile to investigate its mathematical aspects and to try to eliminate certain inconsistencies which it still contains.

2. — The non-orthogonality problem.

Equation (2) contains the so-called «overlap matrix» \mathbf{S} , whose elements must be evaluated for the practical use of the MO-LCAO method. According to its definition (5), \mathbf{S} is in general not zero since the Φ 's centered on different atoms are still to a certain extent overlapping. On the contrary, the different values of the integrals $S_{\mu\nu}$ have been successfully used in the qualitative inter-

pretation of the strengths of different bonds (see *e.g.* COULSON ⁽¹⁾). Notwithstanding this, and other considerations, it has been usually assumed in practical applications of the MO-LCAO method, that the $S_{\mu\nu}$'s could be neglected. This assumption was actually only justified by the extra difficulty of solving the secular equation (2) when overlap was not neglected. However, a large amount of theoretical work was made on the basis of calculations neglecting overlap (as an important example see COULSON and LONGUET-HIGGINS ⁽²⁾). This material could perhaps be utilized in a more strict way, if one could transform an equation including overlap like (2) into an appropriate equation where S no longer appears. This «non-orthogonality problem» became in the long run very important, also because it was realized that neglect of overlap led to very serious difficulties both in applying the MO-LCAO method to chemical problems and in testing the validity of its assumptions; it was solved in a general way, by introduction of «orthogonalized atomic orbitals» (LÖWDIN ⁽³⁾).

For a better understanding of the subsequent derivations, it is useful to report here a few points about this work. LÖWDIN started from equation (2) and found that if the following substitutions are carried out:

$$(6) \quad H' = (1 + S)^{-\frac{1}{2}} H (1 + S)^{-\frac{1}{2}},$$

$$(7) \quad C' = (1 + S)^{\frac{1}{2}} C,$$

(2) becomes

$$(8) \quad H' C' = C' \epsilon.$$

This transformation is actually a transformation of the basis Φ , since one can write (see (3))

$$(9) \quad \Psi = \Phi C = \Phi (1 + S)^{-\frac{1}{2}} C' = \Phi' C'$$

and hence:

$$(10) \quad \Phi' = \Phi (1 + S)^{-\frac{1}{2}}, \quad (\Phi'^{\dagger}, \Phi') = 1.$$

Therefore the transformation just carried out is equivalent to expressing the original operator H_{eff} as a matrix with the new basis Φ' , whose components are what LÖWDIN has called «orthogonalized atomic orbitals». These are suitable linear combinations of the original atomic orbitals. Therefore Löwdin's

⁽¹⁾ C. A. COULSON: *Valence* (Oxford, 1952), p. 198.

⁽²⁾ C. A. COULSON and H. C. LONGUET-HIGGINS: *Proc. Roy. Soc. (London)*, A **191**, 39 (1947); A **192**, 16 (1947).

⁽³⁾ P.-O. LÖWDIN: *Ark. Mat. Astr. Fys.*, A **35**, 9 (1947); *Thesis* (Uppsala, 1948); *Journ. Chem. Phys.*, **18**, 365 (1950).

work can be said to have solved the problem of non-orthogonality by showing that neglect of overlap actually amounts to using a basis which is not built up of strictly atomic orbitals, though this is assumed as a starting point in the LCAO procedure.

Another treatment of the same problem,—which also stressed the importance of overlap in connection with molecules containing heteroatoms,—was given by CHIRGWIN and COULSON ⁽⁴⁾, who analyzed it with special reference to the quantities used in applications of the semi-empirical MO-LCAO method. From the purely numerical point of view, they concluded that the introduction of overlap is not very important for what concerns hydrocarbons, even when one considers polarizabilities. From the general mathematical point of view, the paper in question used an approach which did not imply the transformation of \mathbf{H} into a symmetric matrix \mathbf{H}' as in Lödwin's procedure, but a tensor formalism whose «orthogonalizing» step was equivalent to replace \mathbf{H} of equation (2) by $(\mathbf{1} + \mathbf{S})\mathbf{K}$, with $\mathbf{K} = (\mathbf{1} + \mathbf{S})^{-1}\mathbf{H}$. The matrix \mathbf{K} then fulfilled the equation $\mathbf{K}\mathbf{C} = \mathbf{C}\mathbf{E}$, but was not symmetric. The authors analyzed the possible meaning of the new parameters that were obtained from \mathbf{K} and they suggested that the components of this matrix could be considered as more significant than those of the original matrix \mathbf{H} , at least in connection with electron attracting and electron releasing power induced in atoms and bonds by the geometrical structure of a molecule.

The two works just mentioned did not attempt an explicit connection between the solution of (2) and that of (8), in the sense that their «orthogonalized» matrices \mathbf{H}' and \mathbf{K} were strictly dependent upon the particular structure of the molecule under study, and hence were not directly suitable for the empirical adjustment of parameters which is a basic part of the semi-empirical MO-LCAO procedure.

However, it is possible to solve explicitly the non-orthogonality problem without destroying the one-atom and one-bond dependence of the elements of \mathbf{H} , even in molecules containing heteroatoms, if each overlap integral $S_{\mu\nu}$ is taken proportional to the corresponding bond-integral $\beta_{\mu\nu}$; this suggestion was proposed by MULLIKEN ⁽⁵⁾.

Let us start again from equation (2)

$$(2) \quad \mathbf{H}\mathbf{C} = (\mathbf{1} + \mathbf{S})\mathbf{C}\mathbf{e}.$$

If according to the usual convention, we let α denote the diagonal part of \mathbf{H} , and β denote the matrix $\mathbf{H} - \alpha$, the assumption about overlap we intend to

⁽⁴⁾ B. H. CHIRGWIN and C. A. COULSON: *Proc. Roy. Soc. (London)*, A **201**, 196 (1950).

⁽⁵⁾ R. S. MULLIKEN: *Journ. Chem. Phys.*, **46**, 497 (1949).

introduce here can be written:

$$(11) \quad S = \lambda \beta = \lambda(H - \alpha),$$

λ being an appropriate proportionality factor. Equation (2) then becomes:

$$(12) \quad HC = (1 + \lambda H - \lambda \alpha) C \epsilon,$$

which can also be written as:

$$(13) \quad HC(1 - \lambda \epsilon) = (1 - \lambda \alpha) C \epsilon,$$

thus suggesting the following linear substitution for the diagonal matrix ϵ :

$$(14) \quad \epsilon' = \epsilon(1 - \lambda \epsilon)^{-1}; \quad \epsilon = \epsilon'(1 + \lambda \epsilon')^{-1}.$$

Hence (2) becomes:

$$(15) \quad HC = (1 - \lambda \alpha) C \epsilon'.$$

Eq. (15) is a new eigenvalue equation, similar to equation (2), but with the difference that now, in the place of S , we have a diagonal matrix $-\lambda \alpha$.

Following Löwdin's orthogonalization procedure (equation (6), (7)), let us replace in (15)

$$(16) \quad H' = (1 - \lambda \alpha)^{-\frac{1}{2}} H (1 - \lambda \alpha)^{-\frac{1}{2}},$$

$$(17) \quad C' = (1 - \alpha)^{\frac{1}{2}} C (1 - \lambda \epsilon)^{-\frac{1}{2}}.$$

We thus obtain:

$$(18) \quad H' C' = C' \epsilon',$$

which is the new «orthogonalized» eigenvalue problem.

The importance of the transformation leading from equation (2) to equation (18) lies in the diagonal form of $1 - \lambda \alpha$, since this makes it possible to state explicitly the correlation between each element of H' and the homologous element of H , and between each element of C' and the corresponding element of C .

Let us denote quantities appearing in (18) by the same symbols as those appearing in (2), except for a prime distinguishing quantities of (18) from quantities of (2). We have from (14):

$$(19) \quad \epsilon_i = \frac{\epsilon'_i}{1 + \lambda \epsilon'_i};$$

and from (16)

$$(20) \quad \alpha_{\mu} = \frac{\alpha'_{\mu}}{1 + \lambda \alpha'_{\mu}},$$

$$(21) \quad \beta_{\mu\nu} = \frac{\beta'_{\mu\nu}}{\sqrt{(1 + \lambda \alpha'_{\mu})(1 + \lambda \alpha'_{\nu})}},$$

Equations (20) and (21) provide the possibility, once λ is known, of deriving the parameters α_{μ} and $\beta_{\mu\nu}$ for the non-orthogonal case from those for the orthogonal one, and vice-versa.

As to the eigenvector $\mathbf{C}'_1, \mathbf{C}'_2, \dots, \mathbf{C}'_n$ we shall remark, first of all, that the \mathbf{C}'_i 's are orthogonal and normalized. Supposing \mathbf{C} is normalized according to the standard condition:

$$(22) \quad \mathbf{C}^\dagger(\mathbf{1} + \mathbf{S})\mathbf{C} = \mathbf{1}$$

we have (see eqs. (11) and (17)):

$$(23) \quad \mathbf{C}'^\dagger \mathbf{C}' = \mathbf{1}.$$

Therefore, formula (17), or its equivalent

$$(24) \quad \mathbf{C} = (\mathbf{1} + \lambda \boldsymbol{\alpha}')^{\frac{1}{2}} \mathbf{C}(\mathbf{1} + \lambda \boldsymbol{\epsilon}')^{-\frac{1}{2}}$$

is completely sufficient to define the connection between the orthonormal transformation \mathbf{C}' and the original one \mathbf{C} .

3. - Properties of \mathbf{H} and \mathbf{S} .

Given the entirely formal nature of its derivation, \mathbf{H}' should not be associated with physical quantities (*e.g.* ionization potentials of valence state of atoms, bond energies, etc.) although its elements, according to (20) and (21), retain the one-atom and one-bond dependence characteristic of the elements of \mathbf{H} . As to \mathbf{H} itself, according to a remark by LÖWDIN⁽³⁾, its off-diagonal elements are not invariant under change of the zero-point of potentials, since $\mathbf{S} \neq 0$. This remark in our case is important, because \mathbf{S} depends upon \mathbf{H} , and, therefore, if λ is taken as an invariant quantity, \mathbf{S} is bound to depend upon the zero-point of energies, a result obviously unacceptable. To avoid this difficulty it is necessary to derive a better formulation of \mathbf{H} in eq. (2). Let us introduce a standard reference parameter α , depending on the zero-point the

same way H_{eff} does, and the (invariant) operator

$$(25) \quad \tilde{H}_{\text{eff}} = H_{\text{eff}} - \alpha.$$

Let \tilde{H} be the matrix representation of \tilde{H}_{eff} with respect to the basis Φ , and let $\tilde{\epsilon}$ be its diagonalized form. We have:

$$(26) \quad \tilde{H} = H - (\mathbf{1} + S)\alpha; \quad \tilde{\alpha} =: \alpha - \alpha \cdot \mathbf{1}$$

and since

$$S = \lambda[\tilde{H} + (\mathbf{1} + S)\alpha - \tilde{\alpha} - \alpha \cdot \mathbf{1}] = \lambda(H - \tilde{\alpha}) + \lambda S\alpha$$

we have also:

$$(27) \quad S = \frac{\lambda}{1 - \lambda\alpha} (\tilde{H} - \tilde{\alpha}) = \tilde{\lambda}(\tilde{H} - \tilde{\alpha}).$$

Now, as \tilde{H} is a representation of \tilde{H}_{eff} , it is invariant. Therefore, if $\tilde{\lambda}$ is another invariant, S is actually independent of the zero-point of H_{eff} . In other words, *assumption (11) is acceptable as far as λ is taken as depending upon the zero-point of energies in such a way that, if the zero-point is varied by α , λ varies according to the relation:*

$$(28) \quad \lambda \rightarrow \frac{\lambda}{1 - \lambda\alpha}.$$

The introduction of α and \tilde{H} puts (2) in an invariant form, which has the advantage of making it possible to try to interpret physically the empirical parameters. In fact, by straightforward substitution of (27) and of

$$(29) \quad \epsilon = \tilde{\epsilon} + \alpha \cdot \mathbf{1}$$

into (2) we have:

$$(30) \quad \tilde{H}C = (\mathbf{1} + S)C\tilde{\epsilon}.$$

Since (30) is formally identical to (2), the whole procedure we have described in Section 3 remains valid, and the only actual change concerns the definition (4) of H , which should be replaced by:

$$(31) \quad \begin{cases} \tilde{\alpha}_\mu = \int \Phi_\mu^* H_{\text{eff}} \Phi_\mu d\tau - \alpha. \\ \tilde{\beta}_{\mu\nu} = \int \Phi_\mu^* H_{\text{eff}} \Phi_\nu d\tau - \alpha \int \Phi_\mu^* \Phi_\nu d\tau. \end{cases}$$

4. - Charges and electric dipole moments.

We shall now turn our attention to dipole moments, with special regard to π -dipole moments. We are going to prove that, under a very plausible approximation, dipole moments can be derived from the solution \mathbf{C}' (cf. (17)) of the «orthogonalized» equation (18), hence providing a most natural means to determine the elements of \mathbf{H}' and therefore of \mathbf{H} (see *e.g.* ORGEL *et al.* ⁽⁶⁾, LÖWDIN ⁽⁷⁾). In other words, we can show that the dipole-moment of a π -system can be described by «atomic charges» defined by means of the density matrices \mathbf{P}'_i deriving from (18)

$$(32) \quad \mathbf{P}'_i = \mathbf{C}'_i \mathbf{C}'_i{}^\dagger,$$

that is, just as if (18) were the original equation. Let $\mathbf{R}_i = \mathbf{C}_i \mathbf{C}_i{}^\dagger$ be the i -th density matrix built up from the solution of (2) (LÖWDIN ⁽⁸⁾) we have (cf. (24)):

$$(33) \quad \mathbf{R}_i = \frac{1}{1 + \lambda \varepsilon'_i} (\mathbf{1} - \lambda \boldsymbol{\alpha})^{-\frac{1}{2}} \mathbf{P}'_1 (\mathbf{1} - \lambda \boldsymbol{\alpha})^{-\frac{1}{2}}.$$

If we consider now the electric moment \underline{D}_i , with respect to an origin O , of the charge associated with the i -th molecular orbital obtained from equation (2), we can formulate it (LÖWDIN ⁽⁷⁾) as:

$$(34) \quad \underline{D}_i = e \sum R_{i,\mu\nu} \int \Phi_{\mu\rightarrow}^* \underline{x} \Phi_{\nu} d\tau,$$

where \underline{x} is the distance from O . Let us introduce the following definitions (cf. McWEENY ⁽⁹⁾)

$$(35) \quad \underline{x}_{\mu} = \int \Phi_{\mu\rightarrow}^* \underline{x} \Phi_{\mu} d\tau,$$

$$(36) \quad \underline{s}_{\mu\nu} = \frac{\int \Phi_{\mu\rightarrow}^* \underline{x} \Phi_{\nu} d\tau}{\int \Phi_{\mu}^* \Phi_{\nu} d\tau} - \frac{\underline{x}_{\mu} + \underline{x}_{\nu}}{2},$$

where \underline{x}_{μ} coincides with the distance of atom μ from O , since we are not con-

⁽⁶⁾ L. E. ORGEL, T. L. COTTRELL, W. DICK and L. E. SUTTON: *Trans. Far. Soc.*, **47**, 113 (1951).

⁽⁷⁾ P.-O. LÖWDIN: *Journ. Chem. Phys.*, **21**, 496 (1953).

⁽⁸⁾ P.-O. LÖWDIN: *Journ. Chem. Phys.*, **19**, 1570, 1579 (1951).

⁽⁹⁾ R. McWEENY: *Journ. Chem. Phys.*, **19**, 1614 (L) (1951) and **20**, 920 (1951),
Erratum.

sidering « hybrid » AO 's; $s_{\mu\nu}$ is zero for $\mu = \nu$ and, for $\mu \neq \nu$, it represents the distance of the centroid of the orbitals Φ_μ and Φ_ν from the geometrical centre of the segment $\mu - \nu$. Equation (36) is equivalent to

$$(37) \quad \int \Phi_\mu x \Phi_\nu d\tau = (\delta_{\mu\nu} + S_{\mu\nu}) \left(\frac{x_\mu + x_\nu}{2} + s_{\mu\nu} \right),$$

and hence, by substituting into (34) we obtain:

$$(38) \quad \underline{D}_i = e \sum_\mu [(1 + S)R_i]_{\mu\mu} x_\mu + e \sum_{\mu,\nu} R_{i,\mu\nu} S_{\mu\nu} s_{\mu\nu}.$$

Now, if the Φ 's are very similar to each other, the centroid of Φ_μ and Φ_ν will lie very close to the centre of $\mu - \nu$, and so $s_{\mu\nu}$ can be supposed to be small. Moreover, also the quantities $R_{i,\mu\nu} s_{\mu\nu}$ can be supposed to be much smaller than the diagonal elements of $(1 + S)R_i$.

Therefore, \underline{D}_i can be described in a good approximation as the electric moment of a distribution of point charges

$$(39) \quad Q_{i,\mu\mu} = (Q_i)_{\mu\mu} = [(1 + S)R_i]_{\mu\mu},$$

sitting on the various atoms of the molecule under study:

$$(40) \quad \underline{D}_i \simeq e \sum_\mu Q_{i,\mu\mu} x_\mu.$$

Now, according to (39), (11), and (33), we have:

$$(41) \quad Q_i = \frac{1}{1 + \lambda \epsilon'_i} [(1 - \lambda \alpha)^{\frac{1}{2}} P'_i (1 - \lambda \alpha)^{-\frac{1}{2}} + \lambda (1 - \lambda \alpha)^{\frac{1}{2}} H' P' (1 - \lambda \alpha)^{-\frac{1}{2}}],$$

which, since $H' P'_i = \epsilon'_i P'_i$, transforms into:

$$(42) \quad Q_i = (1 - \lambda \alpha)^{\frac{1}{2}} P'_i (1 - \lambda \alpha)^{-\frac{1}{2}},$$

which was to be expected since R_i must fulfill the relation $R_i \cdot (1 + S) \cdot R_i = R_i$. Now, $(1 - \lambda \alpha)$ is a diagonal matrix, so that the diagonal elements of Q_i will be:

$$(43) \quad Q_{i,\mu\mu} = P'_{i,\mu\mu}.$$

Equation (43), together with (40), enables us to conclude that if n_i is the number of electrons in the i -th molecular orbital, and n_μ the number of electrons shared by the μ -th atom, the total π -dipole moment of the molecule under

consideration is:

$$(44) \quad \underline{M}_{\rightarrow \pi} = e \sum_i n_i \underline{D}_i - e \sum_{\mu} n_{\mu} \underline{x}_{\mu} \simeq e \sum_{i, \mu} (n_i P'_{i, \mu \mu} - n_{\mu}) \underline{x}_{\mu}.$$

Equation (44) expresses the result stated above, namely that, *as far as dipole-moments are concerned*, neglect of overlap in studying π -systems is a plausible approximation, and, in particular, the meaning of « atomic charges » given to the quantities $\sum_i (n_i P'_{i, \mu \mu} - n_{\mu})$ in overlap-neglecting calculations is still substantially justified, even if overlap is taken into account for other purposes. All this, obviously, provided the special assumption (11) for overlap is valid.

5. - First-order perturbation coefficients (« perturbabilities »).

The important work of COULSON and LONGUET-HIGGINS ⁽²⁾ about perturbation theory applied to MO-LCAO calculations led to the definition of a matrix \mathbf{P}_i whose elements are

$$(45) \quad P'_{i, \mu \mu} = \frac{\partial \varepsilon_i}{\partial \alpha_{\mu}}; \quad P'_{i, \mu \nu} = \frac{1}{2} \frac{\partial \varepsilon_i}{\partial \beta_{\mu \nu}}.$$

This matrix, for « orthogonalized » problems, coincides with the density matrix. However, this does not hold when overlap is included (cf. COULSON ⁽⁴⁾). We shall now express \mathbf{P}_i , derived from equation (2), in terms of the eigenvectors \mathbf{C}'_i , derived from (18) and (24). Since (18) does not contain overlap explicitly, the density matrix \mathbf{P}'_i fulfills the relations:

$$(46) \quad P'_{i, \mu \mu} = \frac{\partial \varepsilon'_i}{\partial \alpha'_{\mu}}; \quad P'_{i, \mu \nu} = \frac{1}{2} \frac{\partial \varepsilon'_i}{\partial \beta'_{\mu \nu}}.$$

Now, we have

$$\begin{aligned} \frac{\partial \varepsilon_i}{\partial \alpha_{\mu}} &= \frac{d\varepsilon_i}{d\varepsilon'_i} \left(\sum_{\sigma} \frac{\partial \varepsilon'_i}{\partial \alpha'_{\sigma}} \frac{\partial \alpha'_{\sigma}}{\partial \alpha_{\mu}} + \sum_{\kappa, \tau} \frac{1}{2} \frac{\partial \varepsilon'_i}{\partial \beta'_{\kappa \tau}} \frac{\partial \beta'_{\kappa \tau}}{\partial \alpha_{\mu}} \right), \\ \frac{\partial \varepsilon_i}{\partial \beta_{\mu \nu}} &= \frac{\partial \varepsilon_i}{\partial \varepsilon'_i} \left(\sum_{\sigma} \frac{\partial \varepsilon'_i}{\partial \alpha'_{\sigma}} \frac{\partial \alpha'_{\sigma}}{\partial \beta_{\mu \nu}} + \sum_{\kappa, \tau} \frac{1}{2} \frac{\partial \varepsilon'_i}{\partial \beta'_{\kappa \tau}} \frac{\partial \beta'_{\kappa \tau}}{\partial \beta_{\mu \nu}} \right). \end{aligned}$$

With the help of equations (19), (20), (21), and (46) these expressions can be easily transformed into

$$\begin{aligned} (47) \quad \frac{\partial \varepsilon_i}{\partial \alpha_{\mu}} &= \frac{1}{(1 + \lambda \varepsilon'_i)^2} [(1 + \lambda \alpha'_{\mu})^2 P'_{i, \mu \mu} + \sum'_{\nu} \lambda \beta'_{\mu \nu} (1 + \lambda \alpha'_{\mu}) P'_{i, \mu \nu}] = \\ &= \frac{1 + \lambda \alpha'_{\mu}}{(1 + \lambda \varepsilon'_i)^2} [(1 + \lambda \alpha' + \lambda \beta') \mathbf{P}'_i]_{\mu \mu} = \frac{1 + \lambda \alpha'_{\mu}}{(1 + \lambda \varepsilon'_i)^2} [(1 + \lambda \mathbf{H}') \mathbf{P}'_i]_{\mu \mu}, \end{aligned}$$

and

$$(48) \quad \frac{\partial \varepsilon_i}{\partial \beta_{\mu\nu}} = 2 \frac{\sqrt{(1 + \lambda \alpha'_\mu)(1 + \lambda \alpha'_\nu)}}{(1 + \lambda \varepsilon'_i)^2} P'_{i,\mu\nu}.$$

Consequently, equation (45) becomes

$$(49) \quad P_{i,\mu\mu} = \frac{1 + \lambda \alpha'_\mu}{1 + \lambda \varepsilon'_i} P'_{i,\mu\mu}; \quad P_{i,\mu\nu} = \frac{\sqrt{(1 + \lambda \alpha'_\mu)(1 + \lambda \alpha'_\nu)}}{(1 + \lambda \varepsilon'_i)^2} P'_{i,\mu\nu}.$$

The total perturbabilities are then given by

$$(50) \quad P_{\mu\mu} = \sum_i n_i \frac{1 + \lambda \alpha'_\mu}{1 + \lambda \varepsilon'_i} P'_{i,\mu\mu}; \quad P_{\mu\nu} = \sum_i n_i \frac{\sqrt{(1 + \lambda \alpha'_\mu)(1 + \lambda \alpha'_\nu)}}{(1 + \lambda \varepsilon'_i)^2} P'_{i,\mu\nu}.$$

It must be noted that equations (50) imply a derivation with respect to overlap because of assumption (11).

6. — Discussion.

In order to illustrate the implications of the preceding sections, we shall now consider a few numerical results. For this purpose, the following must be noted:

a) According to the results of Section 3, we can assume that all energies are measured with respect to a given zero-point, which we shall assume as the Coulomb integral of carbon and denote by α . As a unit of measure for energies we shall take as usual the value of the bond integral of a carbon-carbon π bond, and denote this unit by β . With this convention, we can replace the quantities α_μ , $\beta_{\mu\nu}$, ε_i , λ , of the preceding formulae by the new non-dimensional quantities:

$$(51) \quad \delta_\mu = (\alpha_\mu - \alpha); \quad \eta_{\mu\nu} = \beta_{\mu\nu}/\beta; \quad x_i = (\varepsilon_i - \alpha)/\beta; \quad k = \lambda\beta.$$

In the following, when giving values for the various integrals of atoms and bond, and for energies, we shall refer to the quantities defined in eq. (51); as before, we shall distinguish the quantities in question when relating to a calculation neglecting overlap by means of primes, and hence, for the sake of conciseness, we shall call the latter the « primed » parameters, those for the corresponding problem including overlap the « unprimed » parameters.

b) As shown in Section 4, the introduction of overlap into semi-empirical MO-LCAO calculations is pointless as far as electric dipole moments are con-

cerned: therefore, since we shall assume that the empirical fitting of parameters has been carried out via dipole-moments, our starting point will always be a matrix for a problem neglecting overlap; and, since the transformations given above enable us to derive from the solution of this problem the matrix and the solution for a problem which includes overlap and gives the same charge distribution for the system under study, we shall consider the latter as the overlap-including counterpart of the former.

The fact that the fitting procedure can be carried out on the primed parameters does not mean that these are apt to describe changes in a molecular system due to changes in the physical conditions in or around it, at least if the fact that overlap is different from zero is accepted. In particular, if it is intended to find for parameters a correlation with experiment involving a physical interpretation (for instance a correlation of the Coulomb integrals with ionization potentials of atoms, of the bond integrals with the angle of twisting around bonds, etc.) it seems natural to make reference to the unprimed parameters. For example, let us suppose that one tries to evaluate the Coulomb parameters of oxygen and nitrogen by finding those values which lead to correct predicted π -dipole-moments for an appropriate set of molecules (say furan, anisole, pyrrole, etc.). One could, first of all, set up a computation neglecting overlap and, by trial and error, find a satisfactory set of primed parameters. Possible δ' -values for furan oxygen and pyrrole nitrogen could be around 15 and 3 respectively (*). It may appear advisable to introduce corrections for the so-called inductive effect, and this, if considered as a purely heuristic procedure connected with charge distributions, could also be done without taking overlap into account. However, if, at this stage, one wishes to try to correlate the parameters obtained with the ionization potentials of the corresponding atoms, then the quantities to be considered are not the δ' -values but the corresponding δ -values, which are 3.2 and 1.7 respectively (**). This transformation, as can be seen just from the example given, is very important, since, whereas the primed values are very far apart, this is not the case with the unprimed values.

A similar consideration holds also for the η -values. For instance, equation (21) suggests that, if physical considerations suggested that the bond integrals for all π -bonds should be very near to each other, this could not be

(*) We owe this example to Mr. JAN NORDLING, Quantum Chemistry Group of the University of Uppsala, who found that, in order to predict correct dipole moments for the two pairs of molecules, furan and anisole, pyrrole and aniline, parameters of the orders of magnitude given here should be used (if $\eta'_{C-O} > \eta'_{C-N} > \eta'_{C-C}$).

(**) In computing these figures, as in all the results given later, we have assumed that $k = \frac{1}{4}$, since the overlap integral of a CC π -bond is usually taken as 0.25 and η_{CC} is by definition 1.

true of the corresponding primed parameters, unless one actually considers all atomic orbitals orthogonal to each other.

Finally, it must be noted that if we attribute a physical interpretation to the derivatives of the total energy with respect to parameters, we do so by assuming substantially that certain physical phenomena (*e.g.* the approach of a charged particle) change the parameters in question: hence the derivatives to be used must be referred to the unprimed parameters.

c) As has been already mentioned, it is important to remark that the above procedure for the introduction of overlap does not destroy the atom- and bond-correspondence some quantities are given when overlap is neglected. For instance, if the derivative of the total energy with respect to α_μ is associated to atom μ when overlap is neglected, it is still associated to atom μ when overlap is included.

Energies. — As is easily seen from equation (19), the introduction of overlap affects the separation of energy levels though, except for the case (always avoidable by change of the zero-point) when $x'_i < -1/k$, there is no change in distribution with respect to the equivalent overlap-neglecting calculation. The change in separation may involve a difference in the behaviour of the total energy with respect to a change in parameters. Table I shows the example (*) of a substituted benzene PhX, where the primed parameters are $\delta'_x = 2.0$, δ'_o (adj. to X) = 0.2, $\eta'_{cx} = 0.7, 1.0, 1.3, 1.6$. The corresponding unprimed parameters are also given in Table I.

TABLE I. — Total energies of a PhX molecule for (a) a computation neglecting overlap and (b) a computation including overlap and giving the same charge distribution as (a). The Coulomb integral of the substituted Carbon atom is 0.20 for (a) and 0.19 for (b).

$\delta'_x = 2.00$	$\eta'_{cx} = 0.70$	1.00	1.30	1.60
$\delta_x = 1.33$	$\eta_{cx} = 0.56$	0.80	1.04	1.27
(a) $\sum_i n_i x'_i$	12.351	12.503	12.711	12.975
(b) $\sum_i n_i x_i$	8.625	8.605	8.587	8.577

(*) The numerical examples mentioned in this paper are all taken from: G. DEL RE, O. MÅRTENSSON and J. NORDLING (10).

(10) G. DEL RE, O. MÅRTENSSON and J. NORDLING: *A numerical investigation into some aspects of the semi-empirical MO-LCAO method*, Techn. Note 29, Quantum Chemistry Group, Uppsala University, Uppsala.

From Table I it can be seen that the total energy increases when η'_{CX} increases, if computed by neglecting overlap, decreases when η'_{CX} (and hence η_{CX}) increases, if overlap is taken into account. This conclusion is true also of resonance energies, since to evaluate the latter one should subtract from the total energies a constant amount. The peculiar behaviour of the total energy is not due to the fact that we have introduced a correction for «inductive effects», *i.e.* we have given the neighbour of X a Coulomb parameter different from zero. The same behaviour is observed if the Coulomb parameters of all the ring atoms are set equal to 0. However, a disagreement between the two types of calculation is not found over the whole range of possible parameters. The differences in Table I suggest that the energies under (b) tend to reach a minimum and then increase with η_{CX} . This is proven by other calculations we have made: the latter show also that the minimum is reached for a higher value of η_{CX} the higher the Coulomb parameters of X and/or of its neighbours are. This means that for highly electronegative atoms a fairly high repulsion takes place for comparatively low values of η_{CX} : this effect should be more marked if the atom to which X attaches itself has already a high Coulomb integral. For instance—apart from any inclusion of inductive effects,—the formation of a pyridine N-oxide should be, *ceteris paribus*, less easy than that of a trimethylamine N-oxide. This result is obtained only if overlap is taken into account.

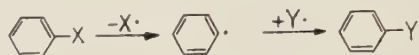
As to inductive effects, these appear to play a comparatively important role. In fact, if the atom adjacent to X is taken as a carbon atom, Table I corresponds to a fairly unrealistic situation. It seems very little plausible that for all values of η'_{CX} the inductive effect should be the same. If we wish to correlate Table I with, for instance, the attack or release of an atom having a $2p\pi$ -lone pair from a substituted benzene, we must consider the inductive effect as subject to a change with η'_{CX} , unless it be neglected: the latter possibility does not seem satisfactory, because it would lead to the conclusion that—from the point of view of the π -system—comparatively highly electro-negative atoms should tend to be repelled by a benzene π -system. One simple possibility would be to assume that the inductive effect is always proportional to the bond parameters η'_{CX} .

If then η'_{CX} is taken as proportional to a reaction co-ordinate, one finds a



Fig. 1.

result which seems quite acceptable. Fig. 1 gives the curve that is obtained when one studies the fictitious process



and attributes to X a $\delta' = 2.0$, to Y a $\delta' = 5.0$, takes $\eta'(\text{CX}) = 1.3$, $\eta'(\text{CY}) = 1.6$ for the stable compounds, and allows for inductive effects by means of the formulae:

$$\delta'(\text{C adj. to X}) = \frac{0.1}{1.3} \eta'(\text{CX}) \delta'(\text{X}); \quad \delta'(\text{C adj. to Y}) = \frac{0.1}{1.6} \eta'(\text{CY}) \delta'(\text{X}).$$

The process in question is also well described by energies obtained without including overlap, even if inductive effects are not taken into account. This implies that, when overlap is not neglected, all results become much more sensitive to changes in the parameters of the atoms of a given π -system, due to the presence of another π -system. Such a remarkable result as that found in Table I does not take place in other simple compounds: for instance, in heterobenzenes, the energy increases steadily with the parameters, both if overlap is included and if it is not. However, the case of substituted benzenes provides a warning against the too hasty conclusion that overlap is unimportant. It is also of interest in connection with the choice of parameters; it shows that, just in the range of primed parameters most often chosen for nitrogen and oxygen (?) the results may be very misleading and that actually either very high or very low values of the parameters may be necessary to obtain satisfactory results.

Molecular diagrams. — The most important consequences of the procedure described in this paper are in connection with those quantities which are usually used to draw the so-called molecular diagrams. It must be emphasized again that we have avoided to introduce or accept new definitions of quantities, but have instead preferred to stick to the standard ones. Atomic charges are those point charges which when assigned to the points where the various nuclei of a molecule are supposed to lie, give a distribution whose dipole moment is practically equal to that of the molecular π -system in question. The perturbabilities are defined according to Coulson and Longuet-Higgins.

In general the introduction of overlap affects the quantities in question enough to make atom and bond perturbabilities sometimes even qualitatively different from charges and bond orders (the latter name being always referred to a calculation neglecting overlap). The simplest example is provided by butadiene. In this hydrocarbon, as in all alternant hydrocarbons, the charges are equal on all atoms. This is not the case when perturbabilities are consi-

dered, since then the two terminal atoms show a strong difference from those of the middle ones (see mol. diagram Fig. 2). This suggests that considerations based on a first-order perturbation procedure are probably enough to serve as a basis for the interpretation of reactions in hydrocarbons. It is also interesting to note that perturbabilities provide, so to say, a compromise between the correlation of atomic reactivities with atomic charges and that, suggested by FUKUI *et al.* ⁽¹¹⁾ with frontier electron densities. In fact, from Eq. (50) it is easy to see that when overlap is introduced, the expression $\sum_i n_i P'_{i,\mu\mu}$ is replaced by $\sum_i n_i m(x'_i) P'_{i,\mu\mu}$ the weight $m(x'_i)$ increasing when x'_i decreases. As to bond-orders, even in the simple case of butadiene they give a picture different from that obtained from total bond perturbabilities. As a matter of fact eq. (50) shows that in the latter the contributions from higher energy levels are even more important than in the case of atom perturbabilities. The results given in Fig. 1b are particularly interesting in connection with the discussion going on at present about the importance of delocalization in connection with bond distances in conjugated polyenes ⁽¹²⁾.

Passing now to the case of compounds containing heteroatoms, we shall consider first of all pyridine-like π systems. The diagram given in Fig. 3 relates to the following choice of « primed » parameters:

$$\delta'_X = 1.0, \quad \eta'_{CX} = 1.3, \quad \eta'_{CC} = 1.0.$$

An « inductive effect » of $\frac{1}{10}\delta'_X$ has been allowed, as in all the computations referred to in this paper, for carbon atoms directly linked to X. The choice of parameters indicated is one made just for the sake of an example, since we

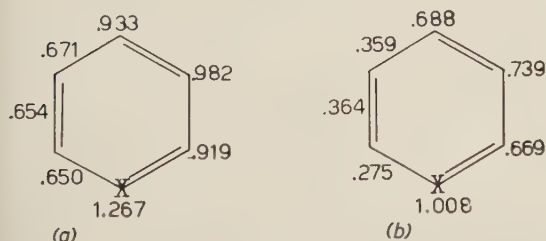


Fig. 3. - Charges and bond-orders (a), and perturbabilities (b) of a heterobenzene, with $\delta'_X = 1.0$ and $\eta'_{CX} = 1.3$.

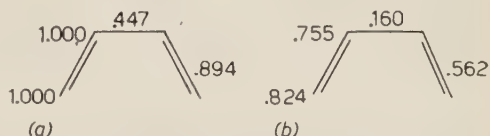


Fig. 2. - Charges and bond-orders (a), and perturbabilities (b) of butadiene.

shall actually make reference, as usual, to computations carried out by changing δ'_X by intervals of 1.0 from 0 to 5, and η'_{CX} by intervals of 0.3 from 0.7 to 1.6.

As is seen from Fig. 3, atom perturbabilities are in close agreement with charges: it is not so of bond perturbabilities and bond orders, since Fig. 2a shows a higher bond order for

⁽¹¹⁾ K. FUKUI, T. YONEZAWA and H. SHINGU: *Journ. Chem. Phys.*, **20**, 722 (1952).

⁽¹²⁾ J. R. PLATT: *Ann. Rev. Phys. Chem.*, **10**, 352 (1954).

the bond 3-4 than for the bond 2-3, while the reverse is true of bond perturbabilities. This may very well be a minor point, since the differences in both cases are very small: hence we can say that the agreement is quite satisfactory. If we neglect very small differences, the same agreement is found over the whole range of parameters; a few additional examples are given in Tables II and III.

TABLE II. - (a) charges and (b) perturbabilities of atoms 2, 3, 4 in the π -systems of pyridine-like molecules.

$\delta'_x \backslash \eta'_{cx}$		0.7		1.0		1.3	
atom		(a)	(b)	(a)	(b)	(a)	(b)
0	2	1.000	.782	1.000	.756	1.000	.732
	3	1.000	.753	1.000	.756	1.000	.758
	4	1.000	.762	1.000	.756	1.000	.751
1	2	.854	.653	.894	.667	.919	.669
	3	.981	.735	.982	.738	.982	.739
	4	.854	.624	.904	.665	.933	.688
2	2	.798	.615	.831	.626	.861	.634
	3	.955	.709	.960	.715	.962	.718
	4	.759	.541	.824	.595	.871	.633

TABLE III. - (a) orders and (b) perturbabilities of bonds 12, 23, 34 in the systems of pyridine-like molecules.

$\delta'_x \backslash \eta'_{cx}$		0.7		1.0		1.3	
bond		(a)	(b)	(a)	(b)	(a)	(b)
0	12	.653	.416	.667	.362	.676	.316
	23	.692	.378	.667	.362	.647	.350
	34	.653	.351	.667	.362	.676	.368
1	12	.573	.311	.624	.299	.650	.275
	23	.710	.400	.678	.379	.654	.364
	34	.639	.332	.659	.349	.671	.359
2	12	.438	.186	.531	.209	.587	.213
	23	.741	.424	.703	.400	.672	.382
	34	.613	.310	.641	.330	.659	.345

However, if quantitative conclusions should be drawn from molecular diagrams, the disagreements, however slight they may be, should perhaps be considered and a decision should be made as to which of the two types of quantities should be used. According to the original papers by COLLSON and LONGUET-HIGGINS⁽²⁾, it seems to us that the use of perturbabilities is probably the most sound procedure in the application of molecular diagrams, especially in connection with bonds: charges retain substantially, according to Section 4, their electrostatic meaning.

Let us consider now another type of molecule, a substituted benzene. As an example (Fig. 4) we can take again the case when $\delta'_x = 2$, $\eta'_{cx} = 1.3$ and as usual, the δ'_c values for carbon atoms are zero, except for that linked to X, whose δ' is 0.2. As is seen from Fig. 4 for the choice of parameters given above the atom perturbabilities are still in a quite satisfactory agreement with charges. The disagreement between bond orders and bond perturbabilities, on the contrary, is so serious that the CX bond has a negative bond perturbability, which may be thought to correspond to something like a repulsive state. This situation is not peculiar to the choice of parameters relating to Fig. 4: it takes place for δ'_x varying from 2 to 5, and for η'_{cx} varying from 0.7 to 1.6. This was expected from the results in Table I, where it was shown that the total energies may decrease with η'_{cx} if overlap is taken into account, contrary to what happens when overlap is neglected. The bond orders are not only of a different sign, but vary in a quite different way with respect to bond perturbabilities. In fact, for a fixed δ'_x the latter first decrease when η'_{cx} —and hence, given the linearity of eq. (22), η_{cx} —increases, and then decreases, whereas the bond-orders seem to be always, so to say, monotonous functions of η'_{cx} (Table IV).

The particular sensitivity of « bond » quantities to the introduction of overlap is due, as we have indicated before, to the fact that we have assumed overlap as depending upon the bond integrals. Atom perturbabilities do not appear to be so sensitive to the introduction of overlap, except when the charges are very near or equal to each other, as in the case of butadiene. It must also be noted that when other quantities, like free-valence indices, are introduced, the importance of overlap is also remarkable, as is easy to understand. In the example of Fig. 4, we find for the valence indices of the *o*-, *m*- and *p*-positions of the substituted benzene mentioned there, the values given in Table V.

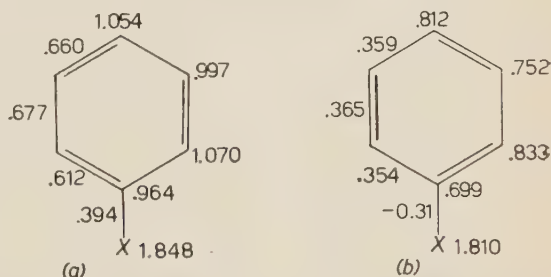


Fig. 4. — Charges and bond-orders (a), and perturbabilities (b) of a substituted benzene with $\delta'_x = 2.0$ and $\eta'_{cx} = 1.3$.

TABLE IV. — (a) orders and (b) perturbabilities of the bond CX in phenol-like π -systems of molecules C_6H_5X .

δ'_X	η'_{CX}	0.7		1.0		1.3		1.6	
		(a)	(b)	(a)	(b)	(a)	(b)	(a)	(b)
0.0	(a)	.501		.635		.728		.792	
	(b)		.280		.332		.353		.355
1.0	(a)	.303		.426		.538		.632	
	(b)		.043		.075		.110		.144
2.0	(a)	.207		.300		.394		.485	
	(b)		— .041		— .042		— .031		— .009
3.0	(a)	.153		.223		.298		.376	
	(b)		— .073		— .093		— .101		— .098

These values indicate an inversion of the ortho-para free-valence indices. This is remarkable, for it has been a common practice to attribute the lower reactivity of the ortho position with respect to the para-position in some sub-

TABLE V. — Valence indices (*) for a calculation neglecting overlap (V') and for one including overlap (V) for the o-, m- and p-positions of a substituted benzene, $\delta'_X = 2.0$, $\eta'_{CX} = 1.3$.

position	o	m	p
V'	.443	.395	.412
V	.489	.484	.491

(*) $V' = 1.732 - \sum_p P'_{\mu\nu}$; $V = 1.208 - \sum_p P'_{\mu\nu}$.

stituted benzenes to steric hindrance: the latter has been used to explain disagreements between calculated and observed reaction rates in the ortho-para substitution of benzene derivatives. Table V provides another possibility for explaining some such disagreements.

7. - Conclusion.

The preceding discussion has tried to examine a few points where the introduction of overlap seems not only to alter the quantitative significance of the various quantities used in applications of the semi-empirical MO-LCAO method, but even to suggest that some correlations to experimental facts should be revised. A few of the latter points we should like to summarize more explicitly.

Suppose we are trying to correlate a schematic ionic reaction—say, with atom quantities as defined above. The introduction of overlap suggests that we should have two effects to take into account: one is the merely electrostatic effect of the charge asymmetry of the molecule, the other is the effect of the actual variation of the energy in the molecule as a consequence of the field produced by the approaching ion. We have for the moment no clear basis on which the two effects could be separated; however, it is possible to assume that, long before an actual modification of the electron distribution in the molecule under study takes place in a non negligible amount, the electrostatic effects become important.

If this is accepted at least as a scheme, we can consider for instance the substitution process at a given position of

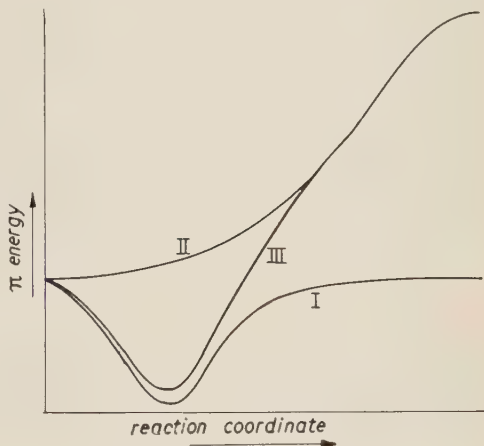


Fig. 5.

a benzene ring as described by an energy curve (III, Fig. 5) which results from that corresponding to an electrostatic interaction which first increases in absolute magnitude with the distance and later tends to become zero (curve I) as a separate quantity, because it is included in the π -electron energy of the entire system (curve II). If we consider an example where the electrostatic effect is an attractive one, we see that there is a possibility of formation of an ionic complex which could even be comparatively stable: this first step would depend mainly upon electrostatic effects and partly upon perturbabilities, whereas the actual formation of an activated complex would depend mainly upon perturbabilities. This type of result is indicative of the modifications (or refinements) in the MO-LCAO description of reactions involving π -systems that are brought about by the results of Sections 4 and 5.

Another point we wish to mention here arises from the remark we have made about the quite common inversion of the predicted ortho-para reactivities that is found when free valence indices are computed by introducing overlap. This has in our opinion a philosophical importance. In fact, the

analysis of physical facts in terms of the simultaneous action of several different « effects » is obviously an artificial procedure, justified as long as these « effects » are clearly defined and in as small as possible a number. Now, the semi-empirical MO-LCAO method in its simplest form has been so successful that it might provide a general basis to build up a semi-empirical theory of organic chemistry in such a way that observed deviations from its predictions might possibly be treated as additional « effects ». However, this requires a very clear analysis of all the details of the method in question. We have made reference for instance to the ortho effect: there is little doubt that the introduction of the latter is useful to explain many experimental facts: but to introduce it as a means of, so to say, correcting the predictions of a « theory » requires having established that the latter cannot possibly give the observed result even if it is more strictly consistent with its own basic scheme. This is essentially the reason why we have considered it worth the while to analyze the « non-orthogonality » problem in the simple MO-LCAO method. Some results, like the more natural way in which the introduction of inductive effects, the importance of frontier electrons, etc., fit with the introduction of overlap give us the impression that, even if many other consequences of the procedure described in this paper (say, transition energies) have still to be analyzed, it may represent a step forward in the sense just outlined.

* * *

The author expresses his gratitude to Dr. P. O. LÖWDIN, under whose guidance most of this work was performed. He wishes also to thank him and the entire Quantum Chemistry Group of Uppsala University for their warm hospitality.

My stay in Uppsala was made possible by the 1958 « C. A. Miranda » Fellowship for Chemistry, for which I wish to thank the Accademia Nazionale dei Lincei (Rome).

RIASSUNTO

Il metodo semi-empirico MO-LCAO viene di solito applicato ai sistemi π trascurando gli integrali di ricoprimento. Nel presente lavoro vengono posti in diretta, esplicita correlazione calcoli in cui si trascurano tali integrali e calcoli in cui essi vengono introdotti sotto la condizione restrittiva che siano proporzionali agli integrali di legame. Diviene così possibile discutere in modo particolareggiato l'effetto dell'introduzione degli integrali di ricoprimento. Per quanto concerne le cariche atomiche, l'introduzione del ricoprimento non produce alcun cambiamento. Tutte le altre quantità vengono invece profondamente modificate, come mostrano sia le formule generali che alcuni esempi numerici. Contrariamente a quanto si è creduto, il trascurare gli integrali di ricoprimento non è lecito in generale, e può influire anche sulla validità quantitativa dei risultati dei calcoli MO-LCAO.

The Levels in ^{121}Sb Studied from the Decay of $^{121}\text{Te}^m$ and ^{121}Te .

R. K. GUPTA (*)

Instituut Voor Kernfysisch Onderzoek - Amsterdam

(ricevuto il 13 Aprile 1960)

Summary. — The levels in ^{121}Sb have been investigated from the decay of ^{121}Te and $^{121}\text{Te}^m$ by single crystal NaI(Tl) spectrometer and coincidence scintillation spectrometer studies. From the coincidences between low intensity 1130 and 70 keV γ -rays and between 506 and 70 keV γ -rays, excited levels at 70, 575, and 1100 keV in ^{121}Sb have been inferred. The new level at 70 keV agrees with the systematics of $\frac{7}{2}^+$ levels in this region.

1. — Introduction.

The decay of 154 day $^{121}\text{Te}^m$ is well investigated ⁽¹⁾: it decays by a cascade of 81.8 keV ($M4$ transition) and 214 keV transitions to the ground states of ^{121}Te . Recently it was reported ^(2,3) that $^{121}\text{Te}^m$ also decays by electron capture to an excited state of ^{121}Sb giving rise to an 1130 keV γ -ray. The 17 day ground state of ^{121}Te is known ⁽⁴⁾ to decay by electron capture to the excited states of ^{121}Sb , giving rise to 506 and 575 keV transitions. A γ -ray of 70 keV

(*) On deputation from Tata Institute of Fundamental Research, Bombay.

⁽¹⁾ Nuclear Data Sheets: NRC 58-4-4, NRC 58-4-1 and NRC 58-3-12 National Research Council, Washington, D.C.

⁽²⁾ K. S. BHATKI, R. K. GUPTA, S. JHA and B. K. MADAN: *Nuovo Cimento*, **6**, 1461 (1957).

⁽³⁾ R. K. GUPTA, S. JHA and B. K. MADAN: *Nuovo Cimento*, **9**, 1117 (1958); see also A. H. WAPSTRA: *Physica*, **19**, 671 (1953).

⁽⁴⁾ M. GOLDBABER and R. D. HILL: *Rev. Mod. Phys.*, **26**, 321 (1954).

has been reported ⁽²⁾ to be in coincidence with 506 keV γ -ray. From these experimental observations the excited states of ^{121}Sb have been placed at 506, 575 and 1130 keV ^(4,2). It was pointed out recently ⁽³⁾ that an examination of the systematics of the occurrence of $d_{\frac{5}{2}}$ and $g_{\frac{3}{2}}$ levels in isotopes of Sb and I shows that a $g_{\frac{3}{2}}$ level in ^{121}Sb should lie a little above the ground state $d_{\frac{5}{2}}$ level and perhaps the 70 keV γ -ray observed in coincidence experiments arises from a level at 70 keV above the ground state, while the 575 keV level decays by 575 and 506 keV transitions to the ground state and 70 keV excited state respectively. If the above assumption is correct the intensity of the 70 keV transition should be at least of equal intensity of the 506 keV transition. Also it may be interesting to investigate the coincidence between the 1130 keV and 70 keV γ -rays to find out if the 1130 keV ray terminates at the 70 keV level and look for a cross-over γ -ray of 1200 keV. Alternately, if the 1130 keV γ -ray represents a ground state transition, a search for the 1060 keV γ -ray connecting the 1130 and 70 keV excited states may be interesting. This investigation was mainly undertaken to throw more light on the low lying $\frac{7}{2}^+$ level in ^{121}Sb predicted from systematics in this region.

2. - Source preparation and γ -ray measurements.

The tellurium isotopes of mass 121 and 123 in ground and isomeric state were produced by bombarding natural antimony with deuterons in the synchrocyclotron of this Institute. The tellurium isotopes were chemically separated from antimony. The tellurium source used for single γ -ray spectrum measurements was about one year old and the 17 day ground state ^{121}Te was in equilibrium with the 154 day ^{121}Te , while a relatively strong source, freshly prepared (≈ 15 days old) was used for coincidence experiments. The single γ -ray spectrum in the region of ≈ 30 keV to 600 keV was measured with a cylindrical NaI(Tl) crystal (25 mm diameter, 25 mm height) coupled to a 6292 Dumont photomultiplier, and the γ -ray spectrum in the region of 100 keV to 1500 keV was taken with a cylindrical NaI(Tl) crystal (63 mm diameter, 62 mm height) coupled to a 6363 Dumont photomultiplier. The spectra were recorded with a 100 channel RIDL analyser. The shape of the 1130 keV photo-peak, was carefully compared with the 1120 keV photo-peak from ^{65}Zn . The close shape fit between the two shows that any other γ -ray 70 keV higher or lower than the 1130 keV γ -ray can not be more intense than 5% of the 1130 keV γ -ray. The γ -ray spectra were analysed by successively peeling off the pulse height distribution due to the various component γ -rays starting at the high energy end. The spectra from sources of ^{203}Hg , ^{85}Sr , ^{137}Cs , ^{114}In , ^{139}Ce and ^{65}Zn were used for energy calibration and also to construct the line shapes of the various peaks resolved in the γ -ray spectrum by interpolation.

The intensities of the γ -rays emitted in the de-excitation of levels in ^{121}Sb fed from the decay $^{121}\text{Te}^m$ and ^{121}Te are summarized in Table I.

TABLE I.

γ -ray energy (keV)	Intensity
70 ± 5	2 ± 1
506	20 ± 3
575	100
1130 ± 10	3 ± 0.6

3. - γ -ray coincidences.

The γ - γ coincidences were measured with a conventional slow-fast coincidence circuit using NaI(Tl) scintillation detectors in both of the channels. The gating peak is selected by a single channel analyser, while the coincidence spectrum in the other channel is displayed by a 100 channel RIDL analyser. The resolving time of the coincidence circuit ($2T$) was ≈ 20 ns. Lead shields covered with silver and copper were used to prevent scattered radiation reaching either detector. Owing to the low intensity of the 1130 keV photopeak, the coincidence rate was low and coincidence runs up to ≈ 12 hours were taken. The low energy γ -ray spectrum in coincidence with the 1130 keV peak from a run of 10 hours is shown in Fig. 1.

The random counting rate was negligible. One can clearly see the anti-mony K -X rays and 70 keV γ -ray in coincidence with the 1130 keV γ -ray. In order to make sure that the 70 keV peak observed in coincidence with the 1130 keV γ -ray does not arise from scattering, the γ -ray spectrum in coincidence with 1130 keV γ -ray from ^{65}Zn taken in the gate was also examined in an identical geometry. The γ -ray spectra in coincidence with 506 and 575 keV γ -rays were also taken in the same geometry. These coincidence studies showed that the 506 keV γ -ray, in agreement with the earlier result (²), is in coincidence with the 70 keV γ -ray and with Sb K -X rays, while the 575 keV γ -ray is only in coincidence with Sb K -X rays. The relatively large intensity of K -X rays in coincidence with the 506 and 1130 keV γ -rays compared with

the 575 keV γ -ray, also shows that K -X rays in coincidence with either 1130 or 506 keV γ -rays are due to K -capture as well as K -conversion.

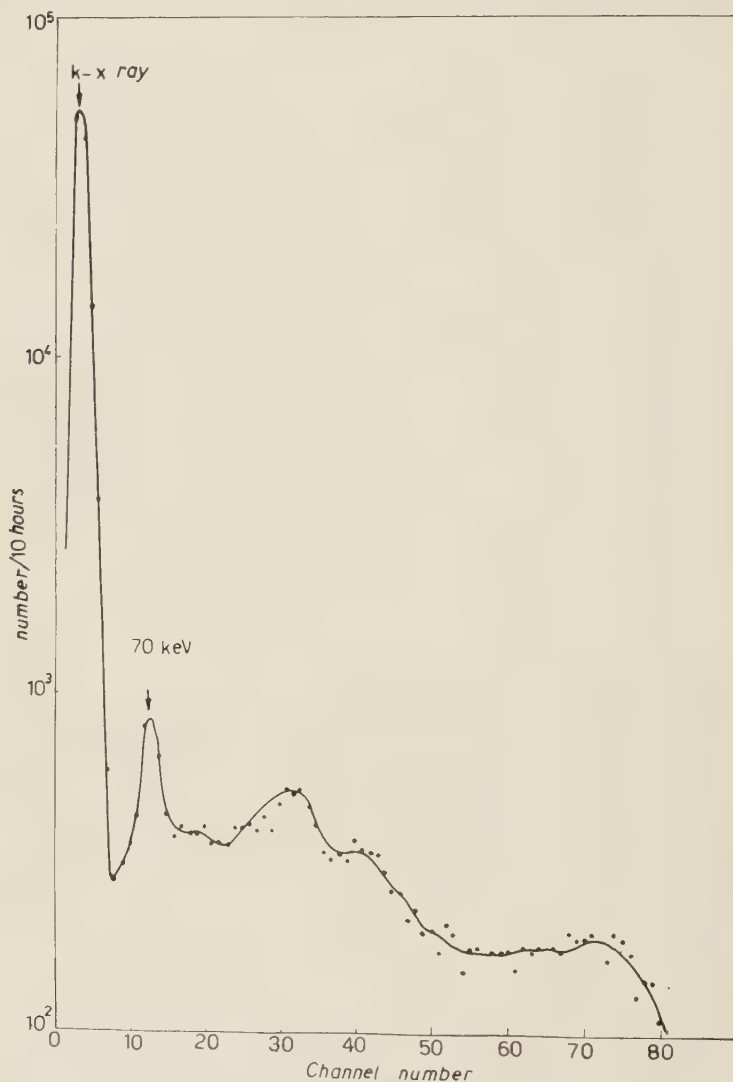


Fig. 1. - Low energy γ -ray spectrum in coincidence with the 1130 keV photopeak

4. - Discussion.

The fact that the 1130 and 506 keV γ -rays are in coincidence with the 70 keV γ -ray, while the 575 keV γ -ray is not in coincidence with any other γ -ray, suggests excited levels at 70, 575 and 1200 keV in ^{121}Sb . A level scheme of ^{121}Sb ,

based on these experimental observations, is given in Fig. 2. The ground state spin of ^{121}Sb can be assigned $\frac{5}{2}^+$ from the measured spin value ⁽⁵⁾ and from shell model considerations. The observance of 70 keV γ -rays in prompt coincidence with 506 and 1130 keV γ -rays shows that the 70 keV transition can be $M1$ or $(M1+E2)$ or $E2$; this is consistent with a $\frac{7}{2}^+$ assignment of the 70 keV level from the systematics. The β^- -decay of the isomeric state of ^{121}Sn with a β^+ group of maximum energy of 420 keV possibly feeds this 70 keV level in ^{121}Sb ⁽⁶⁾. A careful study of the isomeric state of ^{121}Sn will throw more light on this question. The $\log ft$ value

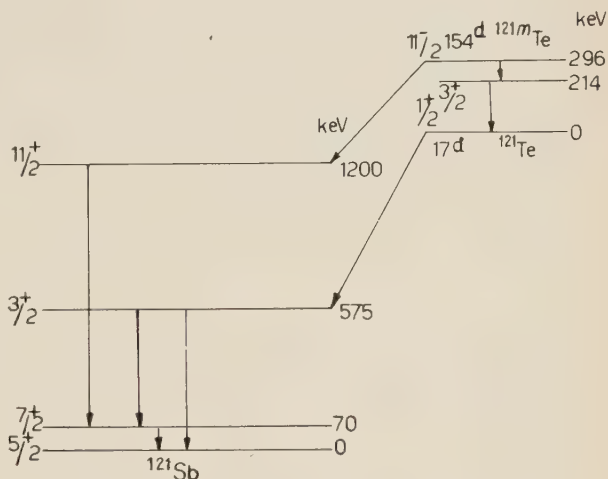


Fig. 2. — Decay scheme of $^{121}\text{Te}^m$ and ^{121}Te .

of the capture branch to the 575 keV level, assuming the value of decay energy ($^{121}\text{Te} \rightarrow ^{121}\text{Sb}$) from Way and Wood's estimate to be 1300 keV ⁽¹⁾, has been calculated ⁽⁷⁾ to be ~ 6.3 .

The assignment of $\frac{1}{2}^+$ ⁽¹⁾ for the ground state of ^{121}Te and the $\log ft$ value of ~ 6.3 for the capture branch to the 575 keV level would suggest an assignment of $\frac{3}{2}^+$ or $\frac{1}{2}^+$ for this state. The experimentally observed ⁽⁴⁾ value of $K/(L+M) \sim 6$ and α (total conversion-coefficient) ~ 0.009 would indicate $M1$ or $E2$ character for the 575 keV transition, consistent with $\frac{3}{2}^+$ or $\frac{1}{2}^+$ assignment of the 575 keV state. The experimentally observed ⁽⁴⁾ value of $K/(L+M) \sim 6$ and $\alpha \simeq 0.018$, comparable intensities of the 506 and 575 keV γ -rays and $\frac{7}{2}^+$ assignment for the 70 keV level would favour an assignment of $\frac{3}{2}^+$ for the 575 keV state. The calculated ⁽⁷⁾ value of $\log ft$ for the capture branch to the 1200 keV state ~ 8.3 ($\log f_i t \sim 7$) and $\frac{11}{2}^-$ assignment for 154 day $^{121}\text{Te}^m$ would suggest $\frac{9}{2}^+$ or $\frac{11}{2}^+$ or $\frac{13}{2}^+$ assignment for 1100 keV state. The non-observance of cross-over transition from the 1200 keV state and the observance of prompt coincidences between the 1130 keV γ -ray and capture K -X rays would favour an assignment of $\frac{11}{2}^+$ for this level.

(5) M. F. CRAWFORD and S. BATESON: *Can. Journ. Res.*, **10**, 693 (1934).

(6) C. M. NELSON, B. H. KETELLE and G. E. BOYD: Oak Ridge National Laboratory Report ORNL-828 (Nov. 1950).

(7) A. H. WAPSTRA, G. J. NIJGH and R. VAN LIESHOUT: *Nuclear Spectroscopy Tables* (Amsterdam, 1959), p. 62.

* * *

The author wishes to thank Prof. Dr. A. H. WAPSTRA and Dr. R. VAN LIESHOUT for their interest in this work and for many useful discussions. He is indebted to Miss J. C. KAPTEIJN for performing the chemical separations and to the personnel of the Synchrocyclotron for the irradiations. This work was performed as part of the research program of the «Stichting Voor Fundamenteel Onderzoek der Materie (F.O.M.)» which is financially supported by the «Nederlandse Organisatie Voor Zuiver-Wetenschappelijk Onderzoek (Z.W.O.)».

RIASSUNTO (*)

I livelli nel ^{121}Sb sono stati analizzati dal decadimento del ^{121}Te e del $^{121}\text{Te}^m$ a mezzo di studi con spettrometro a monocristallo di NaI(Tl) e con spettrometro di coincidenza a scintillazione. Dalle coincidenze fra raggi γ di 1130 e 70 keV a bassa intensità e fra raggi γ di 506 e 70 keV, si sono dedotti livelli eccitati a 70, 575 e 1100 keV nel ^{121}Sb . Il nuovo livello a 70 keV si accorda con la sistematica dei livelli $\frac{7+}{2}$ in questa zona.

(*) Traduzione a cura della Redazione.

On the Fixing of Thick Nuclear Emulsions (*).

I. J. VAN HEERDEN (**) and J. G. McEWEN (**)

Division of Pure Physics, National Research Council - Ottawa

(ricevuto il 26 Aprile 1960)

Summary. — Rate of fixing, clearing time, swelling and corrosion of Ilford G-5 and K-5 emulsions have been studied for different fixing solutions under rigidly controlled conditions. Curves are given which may be used to predict when it is advisable to change some of the fixer.

A satisfactory fixing routine should in a reasonable time give emulsions which are free from unnecessary opacity, corrosion and distortion. Various methods have been used in the past, but in view of the possible wider use of finer grain emulsions, especially for the study of high energy nuclear events, it becomes important to investigate the limitations of nuclear emulsions and methods to overcome them. In order to establish a firmer basis for the various procedures used in fixing, the rate of fixing, clearing times, swelling and corrosion of emulsions were therefore measured for different fixing solutions under rigidly controlled conditions.

To study the effects of the Ag^+ ion concentration on the actual rate of fixing, the following procedure was used: Ilford G-5 and K-5 emulsion pellicles of $600\text{ }\mu\text{m}$ thickness, were cut accurately into pieces of dimensions $1\frac{1}{2}\text{ in.} \times 1\text{ in.}$; these were mounted in pairs of one G-5 and one K-5 emulsion and developed by the normal temperature cycle. Pairs of emulsions were then placed separately in baths containing known quantities of hypo with bisulphite in which various quantities of AgBr had been dissolved. The Ag

(*) Issued as N.R.C. no. 5898.

(**) National Research Council Postdoctorate Fellow. Present address: National Physical Research Laboratory, Pretoria, South Africa.

(**) National Research Council Postdoctorate Fellow. Present address: Laboratory of Nuclear Studies, Cornell University, Ithaca, N. Y.

concentration varied in regular steps from zero to 12 g per litre of fixing solution. The solutions were gently agitated by a continuous rocking of the container and maintained at 6 °C. The liquid in each bath was changed frequently enough to ensure that the Ag concentration never rose above its initial value by more than 0.5 g per litre. At each change a sample of the solution was taken and its Ag concentration determined using a method originally devised by WEYERTS and HICKMAN ⁽¹⁾. In our case, the opacity of the Ag colloid obtained using the buffered solutions given by WEYERTS and HICKMAN, was measured with a Coleman Junior Spectrophotometer. This gave accurate results for concentrations as high as 3 g per litre. For higher concentrations it was necessary to dilute the sample in a known way, and perform the test on the diluted solution; the measurements therefore become progressively less accurate.

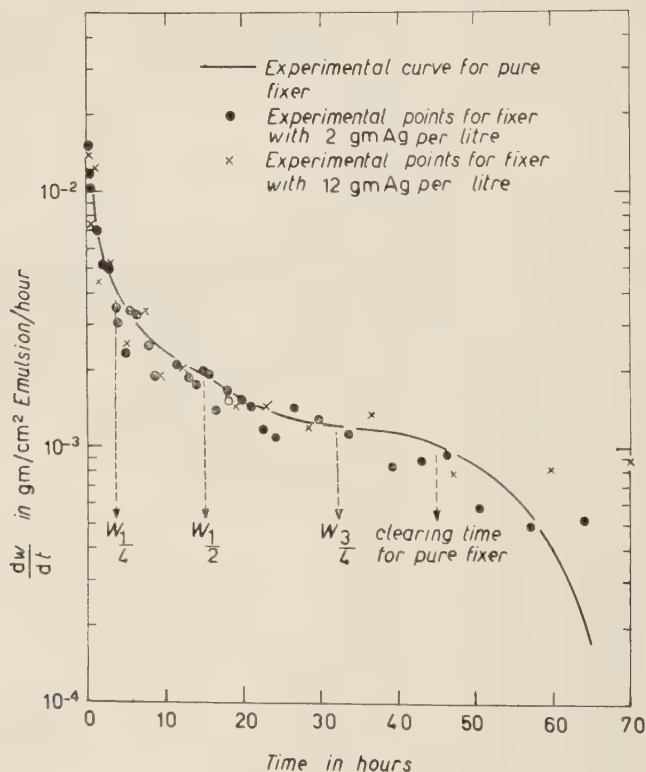


Fig. 1. — Rate of fixing for 600 μ m emulsion in hypo with bisulphite at 6 °C for 0, 2 and 12 g Ag per litre of fixing solution. $W_{\frac{1}{4}}$, $W_{\frac{1}{2}}$ and $W_{\frac{3}{4}}$ denote times for pure fixer at which $\frac{1}{4}$, $\frac{1}{2}$ and $\frac{3}{4}$ of the total silver in the emulsion has been removed.

⁽¹⁾ W. J. WEYERTS and K. C. D. HICKMAN: *Brit. Journ. of Photography*, **82**, 739 (1935).

From these measurements dW/dt , the rate at which silver is removed from the emulsion by fixing solutions with increasing Ag concentrations was determined as a function of time. Typical results are shown plotted in Fig. 1. These all have the same characteristic shape, a very sharp initial decrease in dW/dt , followed by a more gradual one and eventually, after all traces of opaque silver halide have been removed from the emulsion, *i.e.*, when they have cleared, there is a further decrease in the fixing rate. The curve for pure fixer is very well determined; the experimental points for fixing solutions with higher Ag concentrations are less accurate, but they do indicate a slightly lower rate of fixing up to the clearing time of emulsion in pure fixer. In the experimental results of BURGE *et al.* ⁽²⁾ the change in dW/dt is given for the first 20 hours of fixing. Their results are about fifty percent higher than the present results, but this cannot be ascribed to the fact that the Ag concentration of their fixing bath was not kept constant. The rate of fixing does increase with increasing temperature, and the differences might be due partly to the fact that their emulsions were fixed at 8 °C. In this respect it can be mentioned that the results of DENTAN ⁽³⁾ have shown that it is desirable to fix at a low temperature in order to keep distortion at a minimum.

In Fig. 2 the rate of fixing is plotted against percentage of silver removed. It is clear that the variation of rate of fixing is rather complicated and is not well represented by direct proportionality to the amount of silver remaining undissolved. The points for pure fixer are shown in Fig. 2. Within experimental error, those for Ag concentrations up to 12 g per litre are initially the same; the differences become obvious only after about thirty percent of the silver in the emulsion has been removed. The curve shown may therefore be used to predict when changes of fixer are necessary in order to maintain a desired concentration of dissolved silver. To determine the optimum range of Ag concentration it was decided to investigate how the amount of silver in the fixing bath affects clearing time, swelling and corrosion.

Clearing time was defined as the time when visible traces of opaque silver halide could no longer be seen at the bottom of the emulsion. From the results shown in Fig. 2 it seems that in 600 μ m emulsions this occurs when about 90% of the silver has been removed. For the study of the effects of dissolved AgBr on the clearing times, 1½ in. \times 1 in. \times 600 μ m K-5 emulsion pellicles were developed by a normal temperature cycle and fixed in large amounts of fixing solutions, so that the complete removal of silver did not appreciably change the nominal Ag concentration of the fixing solutions. The solutions were stirred and maintained at 6 °C. In addition to the hypo with bisulphite, two

⁽²⁾ E. J. BURGE, J. H. DAVIES, I. J. VAN HEERDEN and D. J. PROWSE: *Nuovo Cimento*, **5**, 1005 (1957).

⁽³⁾ J. P. DENTAN: private communication by Prof. HAENNY.

buffered fixing solutions recently studied by HEUGHEBAERT and HEUGHEBAERT⁽⁴⁾ and one containing hardener were used. The details of the fixing solutions are given in Table I.

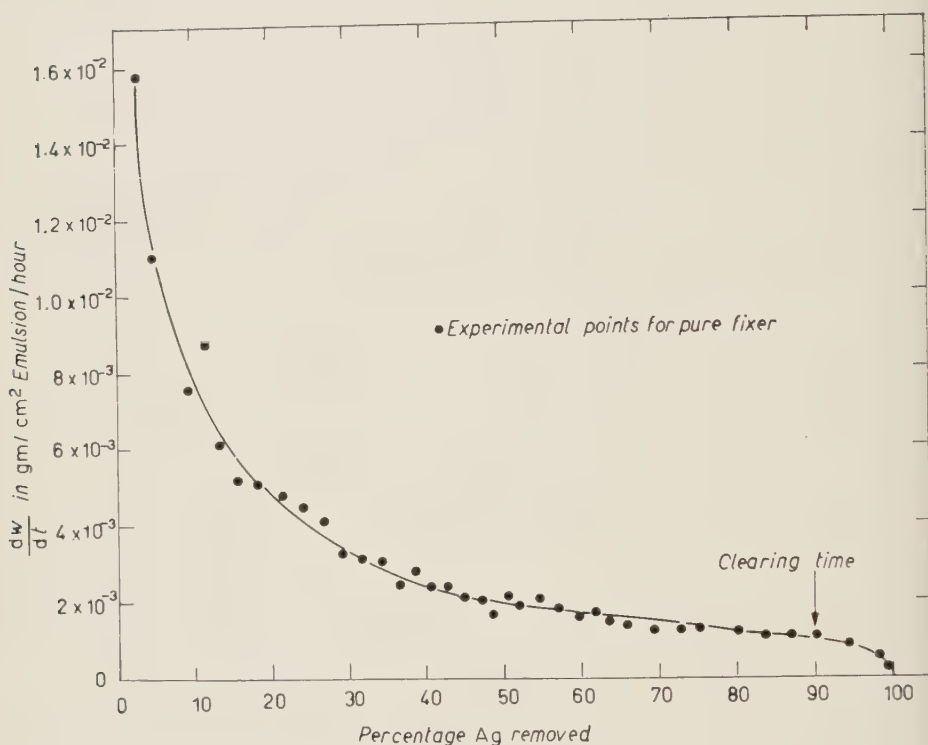


Fig. 2. — Rate of fixing for 600 μ m emulsion in hypo with bisulphite at 6 °C as a function of the percentage silver removed.

Fig. 3 shows how the clearing time for hypo with bisulphite depends on the Ag concentration. It is obvious that with 10 g Ag per litre the clearing time is about fifty percent longer than in pure fixer, and that it increases rapidly with greater concentrations. It is therefore preferable to keep the Ag concentration well below 10 g per litre. The effect of Ag concentration appears to be similar for the other fixing solutions studied but the times are different: an increase in pH decreases the clearing time whereas the addition of hardener increases it. It was also noticed that the yellow toning in the dried emulsion was stronger, the greater the Ag concentration in the fixer.

For the measurements of swelling and corrosion, $1\frac{1}{2}$ in. \times 1 in. \times 600 μ m K-5 pellicles were mounted and given a temperature cycle development whose warm stage at 10 °C consisted of immersion in a boric acid-potassium bromide

(4) D. HEUGHEBAERT and J. HEUGHEBAERT: *Nuovo Cimento*, **12**, 623 (1959).

TABLE I. — *The fixing solutions investigated.*

Hypo with bisulphite	Buffered hypo I	Buffered hypo II	Buffered hypo II with hardener
1	2	3	4
Sodium thio-sulphate 400 g	Sodium thio-sulphate 400 g	Sodium thio-sulphate 400 g	Sodium thio-sulphate 400 g
Sodium meta-bisulphite 30 g	Sodium sulphite 12.5 g	Sodium sulphite 12.5 g	Sodium sulphite 12.5 g
Distilled water 1 litre	Acetic acid (10% solution) 12 ml	Acetic acid (pure) 10 ml	Acetic acid (pure) 10 ml
	Distilled water 1 litre	Distilled water 1 litre	Potassium alum 10 g
			Distilled water 1 litre
pH ~ 5.1	6.8	4.8	4.8

solution: this method has been described in detail by JUDEK ⁽⁵⁾. The thickness of the emulsions was measured during fixing with a mechanical gauge.

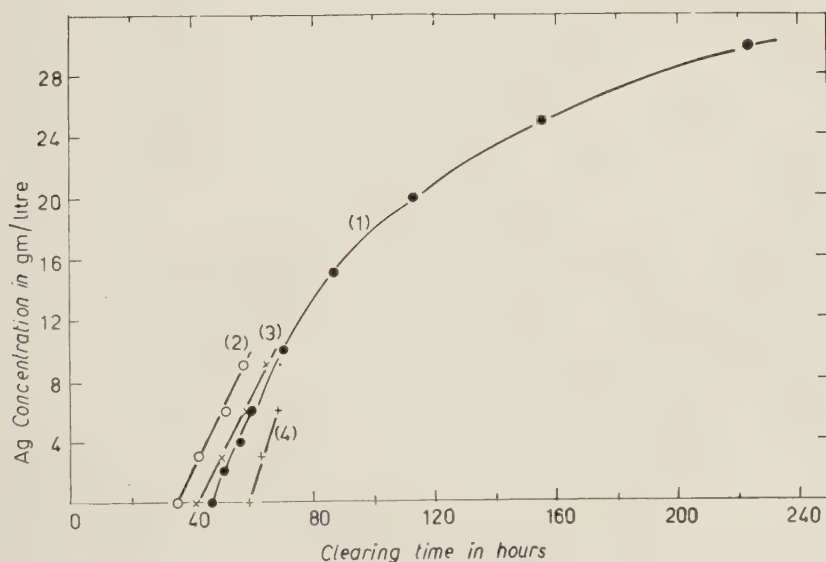


Fig. 3. — Clearing time of 600 μ m emulsion as a function of Ag concentration for various fixing solutions at 6 °C. The numbers on the curves refer to the fixing solutions in Table I.

⁽⁵⁾ B. JUDEK: *Nuovo Cimento*, in press.

The results shown in Fig. 4a were obtained for pure fixing solutions, whereas those in Fig. 4b are for a Ag concentration equal to 4 g per litre. The temperature was again maintained at 6 °C. For all the solutions the maximum thickness was attained around the clearing time. The swelling is followed by

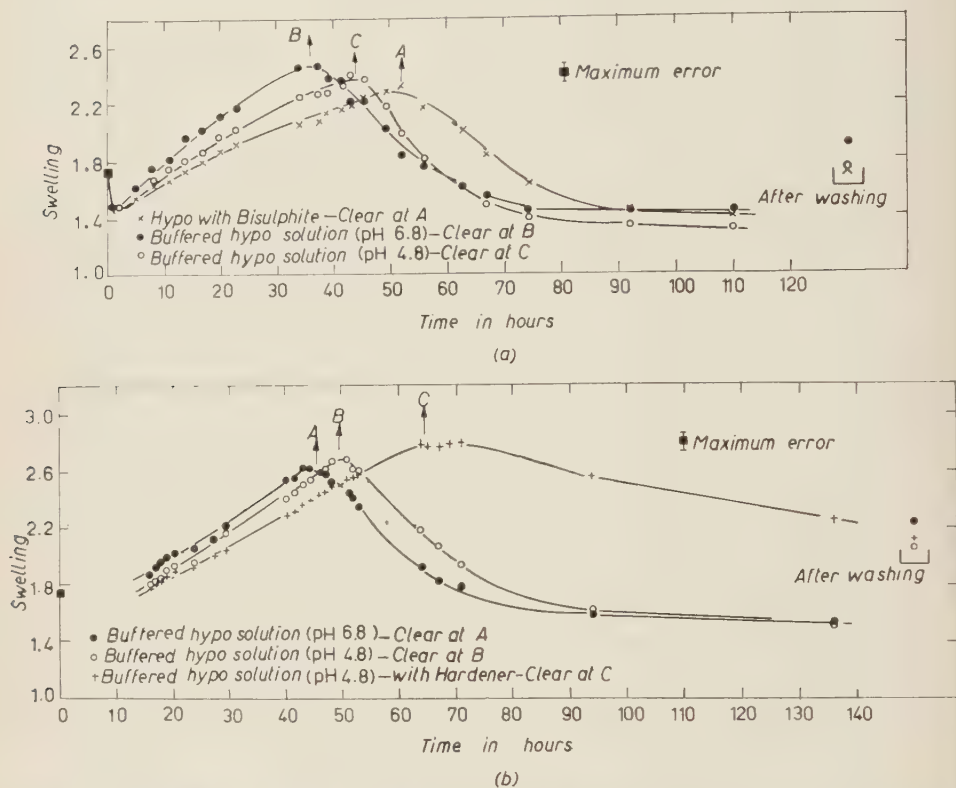


Fig. 4. - Thickness of 600 μm emulsion in fixing bath, temperature 6 °C, divided by original thickness: a) pure fixing solutions and b) fixing solutions with 4 g Ag per litre.

a slightly more rapid shrinking, except in the case of fixer containing hardener where the swelling proceeded rather more slowly and the contraction very much more slowly than with other fixers. The thickness finally attained after washing was considerably greater than that attained after fixing except with fixer containing hardener; in this case the thickness changed only slightly during washing. This suggests that even though the maximum swelling during fixing is slightly higher than with other fixing solutions, the presence of hardener may be advantageous for the minimizing of distortion because of two effects: the swelling takes place at a lower rate and the usual swelling during washing is completely avoided. JUDEK has found that the addition of har-

dener also reduces the number and size of bubbles. It does however also increase the tendency for the stripping of emulsions from the glass, but this may be overcome by soaking the emulsions in a more concentrated glycerine solution than is customary, and by covering them with a transparent coating after drying.

As a measure of the corrosion, the average number of gaps in the last 200 μm of tracks of knock-on protons due to an exposure to 14 MeV neutrons was determined in the K-5 emulsions used for the measurements of swelling. Flat tracks between 40 and 100 μm below the surfaces of the developed plates were selected, and the gap measurements were carried out under very high magnification ($\times 100$ objective and $\times 16$ eyepieces) on a Koristka MS2 microscope. The results for hypo with bisulphite (average $\text{pH} \sim 5.1$) and buffered hypo with $\text{pH} = 4.8$ are shown plotted in Fig. 5. Each point is the mean of 30 tracks. The results show that:

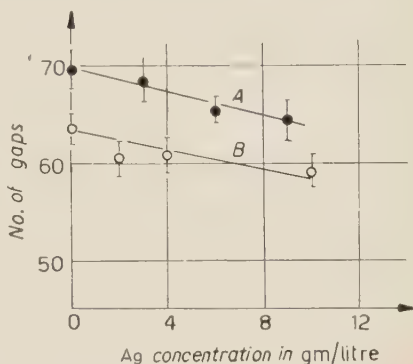


Fig. 5. - Number of gaps in last 200 μm of proton endings in emulsion fixed in A) buffered hypo with $\text{pH} = 4.8$ and B) hypo with bisulphite.

The results for hypo with bisulphite (average $\text{pH} \sim 5.1$) and buffered hypo with $\text{pH} = 4.8$ are shown plotted in Fig. 5. Each point is the mean of 30 tracks. The results show that:

- 1) there is definitely more corrosion in the more acid fixing solution, and
- 2) there is a gradual decrease in the amount of corrosion with increasing Ag concentration.

Both these are in agreement with the results of HEUGHEBAERT and HEUGHEBAERT⁽⁴⁾, who further find that more satisfactory protection against corrosion is obtained by adjusting the pH to 6.8. The results on corrosion do however require very careful checking.

The authors would like to thank Dr. B. JUDEK for many informative discussions.

RIASSUNTO (*)

Si sono studiati il tempo di fissaggio, la durata del lavaggio, il rigontamento e la corrosione delle emulsioni Ilford G-6 e K-5, per diverse soluzioni di fissaggio in condizioni rigidamente controllate. Si danno le curve che possono essere usate per predire quando è conveniente cambiare parte del liquido di fissaggio.

(*) Traduzione a cura della Redazione.

Ghost States and Pair Effects in the Lee Model.

L. M. SCARFONE and W. A. MCKINLEY

Rensselaer Polytechnic Institute - Troy, N. Y.

(ricevuto il 6 Maggio 1960)

Summary. — Pauli and Källén's treatment of the one particle V -states in the ordinary Lee model is applied to J. S. Goldstein's extension of the Lee model which includes pair effects in the θ -particle. Contrary to an earlier conclusion, it is shown that there is simultaneously a ghost state in the V -spectrum with energy greater than the normal V energy as well as a ghost in the θ -spectrum for negative values of one of the renormalization constants.

1. — Introduction.

The Lee model ⁽¹⁾ with pair effects has been studied by GOLDSTEIN ⁽²⁾ by allowing the θ -particle to undergo the transition $\theta \rightleftharpoons \chi + \bar{\chi}$, where χ and its antiparticle, $\bar{\chi}$, are new fermions for this purpose. This transition alone was treated earlier by MACHIDA ⁽³⁾ with results essentially similar to the Lee model. Characteristic of these separate models is the well known fact that normal one particle states can be constructed for renormalized coupling constants less than a critical value which depends upon the cut-off. The squares of the normalization constants of these one particle states lie between zero and one thereby satisfying the criterion for their probabilistic interpretation ⁽⁴⁾. When these theories are extended into the regions beyond their respective critical values these squares become negative thereby leading to difficulties with the

⁽¹⁾ T. D. LEE: *Phys. Rev.*, **95**, 1329 (1954).

⁽²⁾ J. S. GOLDSTEIN: *Nuovo Cimento*, **9**, 504 (1958).

⁽³⁾ S. MACHIDA: *Progr. Theor. Phys.*, **14**, 407 (1955).

⁽⁴⁾ G. KÄLLÉN: *Nuovo Cimento*, **12**, 217 (1954).

normalization of the one particle state. In order to overcome these difficulties it is necessary to introduce an indefinite metric and a ghost state of negative norm and energy less than the normal particle energy. An extension such as this does not eliminate the inherent difficulties as is shown by the non-unitarity of the S -matrix and complex energy eigenvalues⁽⁵⁻⁷⁾. The combined model offers an opportunity to study the behaviour of these ghost states as they occur in a theory with more than one renormalizable field. With respect to this behaviour, a connection between the above mentioned probabilities is developed in reference⁽²⁾ with the result that if the probability associated with the one particle θ -state is negative, that is, a θ -ghost exists in the θ -spectrum then the probability associated with the one particle V -state is necessarily positive and in fact, greater than unity. From this observation it is concluded that the presence of the θ -ghost automatically eliminates the ghost state from the V -spectrum or vice versa. In this paper, we wish to re-examine this behaviour since a study of the states representing a physical V -particle shows that an abnormal state can exist in the V -spectrum when there is indeed a ghost present in the θ -spectrum. This abnormal state has, of course, a negative norm and an energy greater than the normal particle energy.

2. - The combined model and its renormalization (*).

The Hamiltonian for this model is

$$H = H_0 + H_1 + H_2,$$

where

$$(1a) \quad H_0 = \sum_p (M_p - \delta M_p) \psi^*(p) \psi(p) + \sum_p T_p \varphi^*(p) \varphi(p) + \\ + \sum_k (\omega_k - \delta \mu_k) a^*(k) a(k) + \sum_p E_p \chi^*(p) \chi(p) + \sum_p E_p \bar{\chi}^*(p) \bar{\chi}(p),$$

$$(1b) \quad H_1 = g_1 \sum_{pk} X_1(k) [\psi^*(p) \varphi(p-k) a(k) + a^*(k) \varphi^*(p-k) \psi(p)],$$

$$(1c) \quad H_2 = g_2 \sum_{pk} X_2(p) [a^*(k) \chi(k-p) \bar{\chi}(p) + \bar{\chi}^*(p) \chi^*(k-p) a(k)],$$

where $X_1(k) \equiv F_1(k)(2\omega_k\Omega)^{-\frac{1}{2}}$ and $X_2(p) \equiv F_2(p)(2\mu\Omega)^{-\frac{1}{2}}$ with Ω as the normalization volume. The field operators ψ , φ , a , χ and $\bar{\chi}$ are the annihilation operators.

(5) W. PAULI and G. KÄLLÉN: *Dan. Mat. Fys. Medd.*, **30**, no. 7 (1955).

(6) S. WEINBERG: *Phys. Rev.*, **102**, 285 (1956).

(7) R. ASCOLI and E. MINARDI: *Nuovo Cimento*, **14**, 1254 (1959).

(*) We will follow the notation of ref. (2) with minor modifications.

rators for the bare V , N , θ , χ and $\bar{\chi}$ particles, respectively, and obey the usual commutation (anticommutation) relations for boson (fermion) unrenormalized field operators in a Schrödinger momentum representation. The quantities M_p , T_p , ω_k and E_p are the observed energies of these particles and the mass renormalization counter terms for the V and θ -particles are δM_p and $\delta \mu_k$, respectively. The factor $(2\mu\Omega)^{-\frac{1}{2}}$ appearing in $X_2(p)$ replaces the customary $(2\omega_k\Omega)^{-\frac{1}{2}}$ so that one may carry through a renormalization program that is independent of momentum. The observed meson mass is μ . The unrenormalized coupling constants for the transitions $V \rightleftharpoons N + \theta$ and $\theta \rightleftharpoons \chi + \bar{\chi}$ are g_1 and g_2 , respectively, and are related to the renormalized values f_1 and f_2 by

$$(2a) \quad f_1 = g_1 Z_1^{-\frac{1}{2}},$$

$$(2b) \quad f_2 = g_2 Z_2^{-\frac{1}{2}}.$$

$F_1(k)$ and $F_2(p)$ are high momentum cut-off functions for the θ and χ momentum, respectively. The constants of the motion are

$$(3) \quad Q_1 = \sum_p \psi^*(p) \psi(p) + \sum_p \varphi^*(p) \varphi(p),$$

$$(4) \quad Q_2 = \sum_p \varphi^*(p) \varphi(p) - \sum_k a^*(k) a(k) - \frac{1}{2} \sum_p \chi^*(p) \chi(p) - \frac{1}{2} \sum_p \bar{\chi}^*(p) \bar{\chi}(p).$$

Unlike the ordinary Lee model the bare and physical θ -states are no longer identical, however, there is no distinction between bare and physical N 's, χ 's, $\bar{\chi}$'s or vacuum.

The renormalization constant, $Z_2^{-\frac{1}{2}}$, appearing in (2b) is chosen in reference (2) in such a way that it removes a linear divergence from the phase shift expression for χ - $\bar{\chi}$ scattering. However, the scattering still vanishes in the point source limit, $F_2 = 1$, because of a logarithmic divergence which persists in the phase shift expression. To overcome this difficulty it is necessary to retain the cut-off in integrals over χ -momentum. The relation obtained between Z_2^{-1} and f_2^2 is

$$(5) \quad Z_2^{-1} = 1 - (f_2/f_{c2})^2,$$

where the critical constant f_{c2} is defined by

$$(6) \quad f_{c2}^{-2} \equiv \sum_p X_2^2(p) W_{0p}^{-2},$$

and $W_{kp} \equiv E_p + E_{k-p}$. If f_2 is greater than f_{c2} then the probability, Z_2^{-1} , of finding a bare θ in the state of a physical θ is negative as we have remarked in the introduction and as a consequence of the work of PAULI and KÄLLÉN

a ghost state appears in the θ -spectrum. In the presence of a cut-off in the χ -momentum, a logarithmic divergence with respect to θ -momentum which occurs in the \mathcal{N} - θ phase shift expression is eliminated by choosing the renormalization constant $Z_1^{-\frac{1}{2}}$, in (2a), such that

$$(7) \quad Z_1^{-1} = 1 - (f_1/f_{c1})^2,$$

where the critical constant f_{c1} is defined by

$$(8) \quad f_{c1}^{-2} = Z_2^{-1} \sum_k X_1^2(k) D_2^{-2}(k) D_3(k, 0) \omega_k^{-2}.$$

With the exception $D_3(k, 0)$, the D -functions are logarithmically divergent with respect to χ -momentum and are held finite by the above cut-off assumption (*). We observe that if Z_2^{-1} is negative then Z_1^{-1} is positive and greater than unity. However, as we will now show following the procedure of Pauli and Källén, this does not automatically imply the absence of ghosts in the V -spectrum.

3. - The physical V -particle states.

In the following we make the approximation $M_p = T_p = M$, where M is the mass of the \mathcal{N} and physical V -particle. By virtue of the conservation laws (3) and (4) the physical V -state will have the following mixture of bare states all having the same total momentum p_0

$$(9) \quad |V\rangle = \beta [\psi^*(p_0) + \sum_k u(k) a^*(k) \varphi^*(p_0 - k) + \\ + \sum_{kp} v(k, p) \varphi^*(p_0 - k) \bar{\chi}^*(p) \chi^*(k - p)] |0\rangle,$$

where β is a normalization constant and $u(k)$, $v(k, p)$ Fourier coefficients to be determined by the eigenvalue problem. Using (9) with the total Hamiltonian, H , and calling the eigenvalue $M + \omega_0$, we find the equations

$$(10) \quad \omega_0 + \delta M = g_1 \sum_k X_1(k) u(k),$$

$$(11) \quad (\omega_k - \omega_0 - \delta\mu_k) u(k) = -g_1 X_1(k) - g_2 \sum_p X_2(p) v(k, p),$$

$$(12) \quad (W_{kp} - \omega_0) v(k, p) = -g_2 X_2(p) u(k).$$

(*) See appendix for properties of the D -functions.

In the following we restrict ω_0 to the domain $\omega_0 < \mu < 2m$ where m is the mass of γ and $\bar{\gamma}$. Eliminating $v(k, p)$ from (11) and (12) and inserting for $\delta\mu_k$ the expression found in the calculation of the physical θ -state, namely (*),

$$(13) \quad \delta\mu_k = -g_2^2 \sum_p X_2^2(p) (W_{kp} - \omega_k)^{-1}$$

it follows that

$$(14) \quad [1 + g_2^2 \sum_p X_2^2(p) (W_{kp} - \omega_k)^{-1} (W_{kp} - \omega_0)^{-1}] u(k) = -g_1 X_1(k) (\omega_k - \omega_0)^{-1}.$$

The integral appearing on the left of (14) can be cast into the form

$$(15) \quad \sum_p X_2^2(p) (W_{kp} - \omega_k)^{-1} (W_{kp} - \omega_0)^{-1} = \\ = \sum_p X_2^2(p) W_{kp}^{-2} + \sum_p \frac{X_2^2(p) [W_{kp}(\omega_k + \omega_0) - \omega_k \omega_0]}{W_{kp}^2 (W_{kp} - \omega_k) (W_{kp} - \omega_0)},$$

and the first integral on the right of (15) can be written as

$$(16) \quad \sum_p X_2^2(p) W_{kp}^{-2} = \sum_p X_2^2(p) W_{0p}^{-2} + F(k),$$

where $F(k)$ represents the remaining terms of a McLaurin expansion of this integral around $k = 0$. $F(k)$ is proportional to k^2 and finite in the limit $F_2 = 1$. Now using (2b), (5), (14) and (15) we see that (14) becomes

$$(17) \quad u(k) = -g_1 Z_2^{-1} X_1(k) D_1^{-1}(k, \omega_0) (\omega_k - \omega_0)^{-1}.$$

With (17) and (10) we have

$$(18) \quad \omega_0 + \delta M = -g_1^2 Z_2^{-1} \sum_k X_1^2(k) D_1^{-1}(k, \omega_0) (\omega_k - \omega_0)^{-1}.$$

Since $\omega_0 = 0$ must be a solution, it follows that

$$(19) \quad \delta M = -g_1^2 Z_2^{-1} \sum_k X_1^2(k) D_2^{-1}(k) \omega_k^{-1}.$$

The corresponding eigenstate when properly normalized is

$$(20) \quad |V; \omega_0 = 0\rangle = \beta [\psi^*(p_0) - f_1 Z_1^{\frac{1}{2}} Z_2^{-1} \sum_k X_1(k) D_2^{-1}(k) \omega_k^{-1} \varphi^*(p_0 - k) a^*(k) + \\ + f_1 f_2 Z_1^{\frac{1}{2}} Z_2^{-\frac{1}{2}} \sum_{k,p} X_1(k) X_2(p) \omega_k^{-1} D_2^{-1}(k) W_{kp}^{-1} \varphi^*(p_0 - k) \bar{\chi}^*(p) \chi^*(k - p)] |0\rangle,$$

(*) Refer to ref. (2), eq. (13).

where

$$(21) \quad \beta^{-2} = [1 + f_1^2 Z_1 Z_2^{-1} \sum_k X_1^2(k) \omega_k^{-2} D_2^{-2}(k) D_3(k, 0)] .$$

With Källén's ⁽⁸⁾ prescription for the wave function renormalization

$$(22) \quad \langle 0 | \psi_{\text{Ren}}(p_0) | V; \omega_0 = 0 \rangle = 1 ,$$

where $\psi_{\text{Ren}}(p_0) = \psi(p_0) Z_1^{\frac{1}{2}}$, we get

$$(23) \quad \beta^{-2} = Z_1 .$$

Hence, by (21) and (23) we arrive at

$$(24) \quad Z_1^{-1} = 1 - f_1^2 Z_2^{-1} \sum_k X_1^2(k) \omega_k^{-2} D_2^{-2}(k) D_3(k, 0) .$$

(24) agrees with Goldstein's choice of Z_1^{-1} as determined by a study of the \mathcal{N} - θ scattering. The equation for the V -state, (20), becomes

$$(25) \quad \begin{aligned} V; \omega_0 = 0 &= [Z_1^{-\frac{1}{2}} \varphi^*(p_0) - f_1 Z_2^{-1} \sum_k X_1(k) D_2^{-1}(k) \omega_k^{-1} \varphi^*(p_0 - k) a^*(k) + \\ &+ f_1 f_2 Z_2^{-\frac{1}{2}} \sum_{kp} X_1(k) X_2(p) D_2^{-1}(k) \omega_k^{-1} W_{kp}^{-1} \varphi^*(p_0 - k) \bar{\chi}^*(p) \chi^*(k - p)] | 0 \rangle . \end{aligned}$$

We now look for other possible real roots of the eigenvalue problem. Combining (18) and (19) with the aid of (2a) and (24), we get

$$(26) \quad \omega_0 [f_1^{-2} + H(\omega_0)] = 0 ,$$

where

$$(27) \quad H(\omega_0) = Z_2^{-1} \left[\sum_k \frac{X_1^2(k) D_1^{-1}(k, \omega_0) D_2^{-1}(k) D_3(k, \omega_0)}{\omega_k (\omega_k - \omega_0)} - \sum_k \frac{X_1^2(k) D_2^{-2}(k) D_3(k, 0)}{\omega_k^2} \right] .$$

Equation (27) and the properties of the D -functions give

$$(28) \quad H(0) = 0 , \quad \lim_{\omega_0 \rightarrow -\infty} H(\omega_0) = -f_{c1}^{-2} ,$$

$$(29) \quad \begin{aligned} H'(\omega_0) = Z_2^{-1} \left[\sum_k \frac{X_1^2(k) D_1^{-2}(k, \omega_0) D_2^{-1}(k) D_3^2(k, \omega_0)}{\omega_k (\omega_k - \omega_0)^2} + \right. \\ \left. + \sum_k \frac{X_1^2(k) D_1^{-2}(k, \omega_0) D_3'(k, \omega_0)}{(\omega_k - \omega_0)^2} \right] . \end{aligned}$$

⁽⁸⁾ G. KÄLLÉN: *Helv. Phys. Acta*, **25**, 417 (1952).

We note that if $f_2 = 0$ then $Z_2 = D_1 = D_2 = D_3 = 1$ and all the above equations go over into the corresponding equations of the ordinary Lee model as

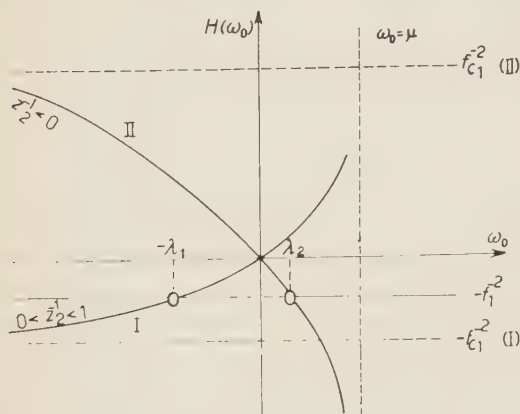


Fig. 1. - Schematic representation of the eigenvalue problem for $f_2 < f_{c2}$ (Curve I) and $f_2 > f_{c2}$ (Curve II). In the former a typical case of $f_1 > f_{c1}$ is shown giving a root to (26) at $-\lambda_1$. As in the ordinary Lee model, this root is not present if $f_1 < f_{c1}$. In the latter case there is a root at λ_2 independent of the relation between f_1 and f_{c1} . We observe that $M - \lambda_1$ takes on any value lying between $-\infty$ and M as f_1 varies from f_{c1} to $+\infty$ and $M + \lambda_2$ takes on any value between $M + \mu$ and M as f_1 varies from 0 to $+\infty$.

normal V -state is present. In addition there now appears a V -state of energy $M + \lambda_2$, $\lambda_2 > 0$ and negative norm (Fig. 1, curve II). In this case there is a normal and θ -ghost present.

4. - Analysis of the norms.

In this section we show that the one particle V -states corresponding to the roots $-\lambda_1$ and λ_2 have negative norms. For this purpose we consider two states of the form (9) with normalization constant $Z_1^{-1/2}B$ and form their scalar product,

$$(30) \quad \langle V; \omega_0 | V; \omega_0 \rangle = Z_1^{-1} B^2 \left[1 + \sum_k u^2(k) + \sum_{kp} v^2(k, p) \right].$$

(*) See Section 4 for discussion of the norms.

Combining (12) and (17) we find the following expression for $v(k, p)$

$$(31) \quad v(k, p) = g_1 g_2 Z_2^{-1} X_1(k) X_2(p) D_1^{-1}(k, \omega_0) (W_{kp} - \omega_0)^{-1} (\omega_k - \omega_0)^{-1}.$$

Substituting (17) and (31) into (30) with the aid of (2a, b), (5) and (24) we find for those values of ω_0 that satisfy $f_1^{-2} = -H(\omega_0)$ that

$$(32) \quad \langle V; \omega_0 | V; \omega_0 \rangle = B^2 f_1^2 \omega_0 H'(\omega_0),$$

as in the ordinary Lee model. Choosing $B = [f_1^2 \omega_0 H'(\omega_0)]^{-\frac{1}{2}}$, then

$$(33) \quad \langle V; \omega_0 | V; \omega_0 \rangle = \frac{\omega_0 H'(\omega_0)}{|\omega_0 H'(\omega_0)|} = \begin{cases} -1, & \omega_0 = -\lambda_1, \\ -1, & \omega_0 = \lambda_2. \end{cases}$$

In (33) we see that in one case the slope is positive and the root negative whereas in the other we have just the opposite. Therefore, these V -states have negative norms and have been normalized to (-1) in (33).

5. - Conclusions.

We have seen that the extended Lee model with pair effects in the θ -particle has the possibility of three stable mass states in the V -spectrum. One of these is the normal particle state of energy M and the other two are abnormal states. Of the latter two, both have negative norms but one has energy less than the normal particle energy and the other has energy greater than the normal particle energy. As for the first of these abnormal states, we recognize its similarity to the V -ghost which appears in the ordinary Lee model. For example, for fixed $f_2 < f_{c2}$, the energy of this state lies between $-\infty$ and M for values of f_1 between f_{c1} and $+\infty$ ⁽⁹⁾. If the pair effects vanish ($f_2 = 0$) this state coincides with the Lee V -ghost. The second abnormal state mentioned above has a different behaviour under variation of f_1 for fixed $f_2 > f_{c2}$. The energy of this state can range between M and $M + \mu$ as f_1 ranges between $+\infty$ and 0 . The unphysical character of these abnormal states is clear.

It appears then that both renormalized fields in this model can simultaneously contain a ghost state and this occurs when one of the probabilities (Z_2^{-1}) is negative and the other (Z_1^{-1}) greater than unity.

(9) Y. MUNAKATA: *Progr. Theor. Phys.*, **13**, 455 (1955).

APPENDIX

D-functions.

These functions are defined by

$$(A.1) \quad D_1(k, \omega_0) = D_3(k, 0) + f_2^2 \sum_p \frac{X_2^2(p) [W_{kp}(\omega_k + \omega_0) - \omega_k \omega_0]}{W_{kp}^2 (W_{kp} - \omega_k)(W_{kp} - \omega_0)},$$

$$(A.2) \quad D_2(k) \equiv D_3(k, 0) + f_2^2 \sum_p \frac{X_2^2(p) \omega_k}{W_{kp}^2 (W_{kp} - \omega_k)},$$

$$(A.3) \quad D_3(k, \omega_0) = D_3(k, 0) + f_2^2 \sum_p \frac{X_2^2(p) \omega_0}{W_{kp}^2 (W_{kp} - \omega_0)},$$

$$(A.4) \quad D_3(k, 0) \equiv 1 + f_2^2 F(k).$$

They are connected according to the relation

$$(A.5) \quad D_1(k, \omega_0)(\omega_k - \omega_0) + \omega_0 D_3(k, \omega_0) = \omega_k D_2(k).$$

For fixed k , the derivatives of D_1 and D_3 with respect to ω_0 are

$$(A.6) \quad D_1'(k, \omega_0) = f_2^2 \sum_p \frac{X_2^2(p)}{(W_{kp} - \omega_k)(W_{kp} - \omega_0)^2},$$

$$(A.7) \quad D_3'(k, \omega_0) = f_2^2 \sum_p \frac{X_2^2(p)}{W_{kp} (W_{kp} - \omega_0)^2}.$$

Lastly,

$$(A.8) \quad \lim_{\omega_0 \rightarrow -\infty} D_1(k, \omega_0) = \lim_{\omega_0 \rightarrow -\infty} D_3(k, \omega_0) = Z_2^{-1}, \quad k \text{ fixed.}$$

RIASSUNTO (*)

Il trattamento di Pauli e Källén per gli stati V ad una particella nel modello ordinario di Lee viene applicato alla estensione di J. S. Goldstein del modello di Lee, la quale include gli effetti delle coppie sulla particella θ . In contrasto con una conclusione precedente, si mostra che si hanno simultaneamente uno stato fantasma nello spettro V con energia più grande dell'energia V normale ed un fantasma nello spettro θ per valori negativi di una delle costanti di rinormalizzazione.

(*) Traduzione a cura della Redazione.

The Thermal Neutron Activation Cross Section of ^{105}Ru .

B. L. SHARMA (*)

Department of Physics, Ohio State University - Columbus, Ohio

(ricevuto il 10 Maggio 1960)

Summary. — In order to calculate the thermal neutron activation cross section of ^{105}Ru , the enriched ^{104}Ru was exposed to a thermal neutron flux of $1.2 \cdot 10^{14}$ neutrons $\text{cm}^{-2} \text{s}^{-1}$. After the decay of the 40-day activity of ^{103}Ru , the silver impurity was removed and the contributions from Co and Eu impurities were subtracted. The γ -ray energies and the coincidence results were consistent with the previously established results for ^{106}Ru - ^{106}Rh . From γ -ray intensities consideration, the thermal neutron activation cross section of ^{105}Ru was calculated to be (0.20 ± 0.02) barn.

1. — Introduction.

The γ radiation from ^{106}Rh , a 30 s β -activity ⁽¹⁾ which is generally used in an equilibrium condition with 1 year ^{106}Ru , has been studied extensively during the past few years. The first complex decay scheme of ^{106}Rh was proposed by PEACOCK ⁽²⁾ in 1947, and the first experimental evidence for greater complexity of the ^{106}Rh decay scheme was obtained by GOLDHABER and DER MATEOSIAN ⁽³⁾ in 1950, who discovered that this activity could produce photo-neutrons in Be and D_2O .

(*) Present address: Defence Science Laboratory, Delhi, 6.

(1) L. D. GLENDENIN and E. P. STEINBERG: *National Nuclear Energy Series, Plutonium Project Record*, **9**, 793 (1951).

(2) W. C. PEACOCK: *Phys. Rev.*, **72**, 1049 (1947).

(3) M. GOLDHABER and E. DER MATEOSIAN: *Brookhaven National Laboratory Report BNL*, **51**, S-5 (1950).

The ^{106}Rh decay and the energy levels for the excited states of ^{106}Pd have been suggested by HAYWARD (⁴) and ALBURGER (⁵). Later γ radiations of 30 s ^{106}Rh have been examined by KAHN and LYON (⁶) by using a NaI(Tl) γ -ray spectrometer and the decay scheme proposed by them is essentially in agreement with the one proposed by ALBURGER except for an additional level in ^{106}Pd at 2.28 MeV. In all the above mentioned investigations, Ru separated from fission products was used as a source for the measurement of the decay of ^{106}Rh .

In the present investigation, an attempt was made to identify the γ radiations from ^{106}Rh from a completely different angle. The enriched isotope ^{104}Ru was used to investigate ^{106}Ru and consequently ^{106}Rh , which can be formed from ^{104}Ru by two consecutive neutron captures. The decay scheme proposed by ALBURGER (⁷) and a $^{106}\text{Ru} - ^{106}\text{Rh}$ source obtained from Oak Ridge National Laboratory, were used as standards for identifying and comparing the activity of ^{106}Rh . After identifying this activity an attempt was made to estimate the thermal neutron activation cross-section of ^{105}Ru .

2. - Experimental procedure.

Fifteen milligrams of ruthenium enriched in isotope ^{104}Ru (98.2%) was exposed to a thermal neutron flux of $1.2 \cdot 10^{14}$ neutrons $\text{cm}^{-2} \text{ s}^{-1}$ for 176.8 hours at the Materials Testing Reactor of the Phillips Petroleum Company, Idaho. After the irradiation, the γ radiations of this sample were examined by using the scintillation detectors, which were composed of $1\frac{1}{4}$ in. \times 2 in. cylindrical NaI(Tl) crystal, a DuMont 6292 photomultiplier tube, and a white cathode follower designed at Oak Ridge National Laboratory. The pulses from this type of detector were fed into 100 channel pulse height analyzer.

At the beginning of the measurements, the activity of this sample was very high and was mainly due to 39.7 day ^{103}Ru . The decay of 498 keV γ -ray was followed on a single channel pulse height analyzer and was observed to decay with a half-life of 40 days. Because of this ^{103}Ru activity, which was formed in sufficient amount to mask any activity of ^{106}Ru (half-life 1 yr), this sample was allowed to decay for a period of 400 days. After this period, it was observed that the activity of ^{106}Ru was still masked by the activity of ^{110}Ag (half-life 270 days), which was present in the sample as an impurity. The

(⁴) R. H. HAYWARD: *Phys. Rev.*, **86**, 760 (1952).

(⁵) D. E. ALBURGER: *Phys. Rev.*, **88**, 339 (1952).

(⁶) B. KAHN and W. S. LYON: *Phys. Rev.*, **92**, 902 (1953).

(⁷) D. STROMINGER, J. M. HOLLANDER and G. T. SEABORG: *Table of Isotopes* (University of California, Radiation Laboratory Report, 1958).

chemical separation of the radioactive silver from this sample was done as follows. In order to dissolve ruthenium without spattering, it was sealed together with a fixed amount of concentrated HNO_3 and HCl in a glass tube which was heated in a furnace at constant temperature. After it was dissolved, the silver fraction, with the addition of silver carrier, was precipitated by the standard procedure used for precipitation of silver. The method of precipitation was used again and again for a number of times in order to remove the radioactive silver (^{110}Ag) completely.

After the silver impurity was removed completely, and the contributions from Co and Eu impurities were subtracted, the 513 and 621 keV γ -rays of ^{106}Rh were observed. In order to identify more conclusively the origin of 513 and 621 keV photo-peaks, coincidence measurements were made. The γ - γ coincidences were observed by placing this sample between two scintillation heads, each containing a $1\frac{3}{4}$ in. \times 2 in. NaI(Tl) crystal. In the first γ - γ coincidence measurements, the single channel analyzer was gated to accept the pulses corresponding only to 513 keV γ -ray, while the 100 channel pulse height analyzer viewed the entire spectral region. This measurement showed that 513 keV γ -rays was in coincidence with the 621 keV γ -ray. These measurements were also compared with those of the standard ^{106}Ru - ^{106}Rh source obtained from Oak Ridge and were found to be in agreement.

3. - Results and conclusions.

After verifying the activity of ^{106}Rh , the thermal neutron activation cross-section of ^{105}Ru was calculated from γ -ray intensity considerations. Knowing the crystal efficiency of $1\frac{3}{4}$ in \times 2 in NaI(Tl) crystal ⁽⁸⁾, it was possible to calculate the number of radioactive atoms of ^{106}Rh at any time t after irradiation. Using 513 keV γ -ray intensity and the thermal neutron activation cross-section of ^{104}Ru ($\sigma = 0.7$ barn) ⁽⁹⁾, the thermal neutron activation cross-section of ^{105}Ru was calculated to be 0.20 barn. Whereas the thermal neutron activation cross-section of ^{105}Ru , calculated from 621 keV γ -ray intensity consideration was found to be 0.24 barn. Although the two calculated values of the cross-section are in close agreement with each other, there would be some error present in them due to the activities produced from the impurities. The question of impurities is very important because the successive neutron capture method is a second order process. It is true that scintillation spectrometer

⁽⁸⁾ E. A. WOLICKI *et al.*: *Calculated efficiencies of NaI crystal*, NRL Report 4883, 32 (1956).

⁽⁹⁾ W. H. SULLIVAN: *Trilinear chart of nuclides*, AEC (Jan. 1957).

and coincidence counting methods reduce the error caused by the activities produced from the impurities, but at the same time they bring forward such problems as a quantitative knowledge of the branching ratios or the intensity percentage values whenever more than one γ -ray is emitted.

* * *

The author wishes to thank Professor M. L. POOL for suggesting this problem and for his interest and encouragement throughout this work.

RIASSUNTO (*)

Allo scopo di calcolare per il ^{105}Ru la sezione trasversale di attivazione ai neutroni termici, il ^{104}Ru arricchito fu esposto ad un flusso di neutroni termici di $1.2 \cdot 10^{14}$ neutroni $\text{cm}^{-2} \text{s}^{-1}$. Dopo il decadimento della attività di 40 giorni del ^{103}Ru , l'impurità di argento venne rimossa ed i contributi delle impurezze di Co ed Eu vennero sottratte. Le energie dei raggi γ ed i risultati delle coincidenze risultarono in accordo con dati precedentemente raccolti per ^{106}Ru - ^{106}Rh . Dall'analisi delle intensità dei raggi γ , si è calcolato che per il ^{105}Ru la sezione trasversale di attivazione ai neutroni termici è di (0.20 ± 0.02) barn.

(*) Traduzione a cura della Redazione.

Search for γ Radiation from the Cygnus A Radiosource.

A. BRACCESI and M. CECCARELLI

Istituto di Fisica dell'Università - Bologna

G. SALANDIN

Istituto Nazionale di Fisica Nucleare - Sezione di Padova

(ricevuto il 11 Maggio 1960)

Summary. — An experiment is described attempting to detect energetic γ -radiation from a peculiar celestial object (Cygnus A) interpreted by some authors as a collision between a galaxy of matter and one of anti-matter. The experiment was negative and settled as upper limit for the γ -flux a value of about $5 \cdot 10^{-2}$ quanta/cm² s, which is about two orders of magnitude smaller than estimated by Morrison under the assumption previously mentioned.

1. — Introduction.

In several occasions the hypothesis has been advanced that in nature might exist comparable amounts of matter and anti-matter, the two forms being independently aggregated in large bodies having very little mutual interaction so to allow the essential stability of the universe.

The physical properties of the two forms of matter being of course identical the only chance of testing this hypothesis would be either to obtain actual samples of extragalactic matter, as for instance extreme energy cosmic rays, or to have the chance of observing the almost certainly unconventional features of collisions eventually occurring somewhere in the sky between two large aggregates of matter and anti-matter.

The two major phenomena observable in such a collision, pointed out respectively by HOYLE and BURBIDGE ⁽¹⁾ and by MORRISON ⁽²⁾ are:

⁽¹⁾ F. HOYLE and G. R. BURBIDGE: *Nuovo Cimento*, **4**, 558 (1956).

⁽²⁾ P. MORRISON: *Nuovo Cimento*, **7**, 858 (1958).

a) Emission of fast electrons through the chain $N + \bar{N} \rightarrow \pi \rightarrow \mu \rightarrow e$ and hence, in presence of magnetic fields, strong synchrotron radio emission.

b) Emission of soft γ -rays from electron-positron annihilation and of γ -rays in the vicinity of 100 MeV from π^0 decay.

Were the emission of radio or soft γ quanta may be originated in a number of other phenomena, energetic collimated γ 's would be almost unmistakable fingerprints of an annihilation process.

Because of the concomitance of processes a) and b) the celestial objects *a priori* better suited for a search of annihilation γ -radiation are the strong and highly brilliant extragalactic radio-sources; the leader of this class for the terrestrial sky being the Cygnus A source.

The very high radio brilliance of this object, the accurate correspondence between its radio co-ordinates and the optical ones of an object interpreted by BAADE and MINKOWSKY ⁽³⁾ as two closely colliding galaxies and, finally, statistical considerations on the distribution of radiosources of the same class of brilliance strongly point out that the Cygnus source is indeed a highly peculiar encounter of two galaxies.

2. - γ -flux estimates and experimental results.

Estimates of the γ -flux in case the Cygnus A source were the collision between a galaxy and an anti-galaxy have been done by MORRISON ⁽²⁾ and by SAVEDOFF ⁽⁴⁾.

The former, in the hypothesis of an head-on collision of two clouds of gases of essentially known mass and velocity ⁽³⁾ estimates a flux at the top of our atmosphere of about 1 γ quantum/cm² s. The latter assumes that about the same amount of energy feeds the radio and the γ channels or, in other words, that magnetic fields intensities and gas densities are such to allow practically full conversion of the fast annihilation electrons energy into radio waves. From the experimental value of the energy flux of this channel one gets for the γ -flux the much discomforting value of $5 \cdot 10^{-7}$ quanta/cm² s; a value quite outside the present experimental possibilities.

In the experiment we are going to describe we have not detected δ radiation from the Cygnus source up to a limit of $5 \cdot 10^{-3}$ quanta/cm² s; well outside the Morrison estimate. For those who like to maintain the hypothesis that in the source matter and anti-matter are interacting, our experiment points

⁽³⁾ W. BAADE and R. MINKOWSKY: *Astrophys. Journ.*, **119**, 206 (1954)

⁽⁴⁾ M. P. SAVEDOFF: *Nuovo Cimento*, **13**, 12 (1959).

out the opportunity of postulating some sort of magnetohydrodynamical process able to prevent the two clouds of interstellar gas to undergo full contact.

3. — Details on the experiments.

A little stack of Ilford G-5 Nuclear Research Emulsions has been flown one hour at an altitude of 27 km (17 g/cm^2) at the time of culmination of the Cygnus source. Since this source has a declination of $+40.5^\circ$, not very different from the latitude of the site of the experiment (44.5°), a collimated γ -beam will lead, in spite of rotations of the balloon, to electron pairs all forming angles of less than 7 degrees with the vertical of the stack.

In order to reduce background due to the soft component of cosmic radiation the emulsions were made to turn automatically by 180 degrees on a horizontal axis after a time corresponding to the ceiling of the balloon, this being predetermined to occur half hour before the culmination of the source, and again by the same amount after one hour at the time of release. In this way the plates had the proper orientation only during the high altitude part of the flight and hence were not contaminated by ground level or intermediate altitude electron pairs, except for albedo.

In addition to the mechanism for rotating the plates the gondola contained a mercury barometer and a thermometer with continuous recording as well as a device which, by taking a sort of picture of the sun ensured that the equipment was not swinging significantly.

The equipment had a total weight of 2 kg and was flown by means of a neoprene Darex 1750 g balloon fitted with a release valve ⁽⁵⁾. The bal-

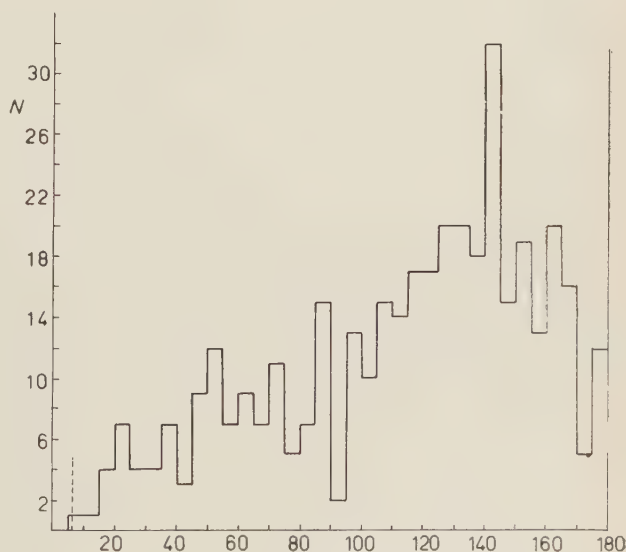


Fig. 1. — Number of electron pairs as a function of the zenithal angle. The dotted line limits the angular region in which γ 's from the Cygnus A were expected.

⁽⁵⁾ J. E. LABY, Y. K. LIM and V. D. HOPPER: *Nuovo Cimento*, 5, 249 (1957).

loon performance for day flights was excellent; we were also able to secure exposures at night, although with less reliability because of the increased fragility of balloons.

In Fig. 1 is plotted the angular distribution of the electron pairs referred to the vertical of the stack. The dotted line at the left of the figure limits the region within which was expected the radiation from the Cygnus source. The fact that the majority of the γ particles do apparently enter from the bottom of the stack ($\theta > 90^\circ$) shows the effectiveness of the procedure used for reducing the background.

The results of a double scanning have shown that, at least for flat electron pairs of an energy around 100 MeV, the loss is not likely to be larger than 50 %. By taking into account the scanning loss, the matter above the plates, the conversion mean free path, etc., it results that one electron pair corresponds to a flux of $5 \cdot 10^{-3}$ γ quanta/cm²s. About this value is then the sensitivity of our experiment and hence the upper limit we can give for γ -radiation from the Cygnus A radiosource.

* * *

Professor PUPPI is cordially acknowledged for his suggestions and encouragement, Professors MANNINO and QUARENI and Mrs. STAGNI and TONNI for help during the flights, Mr. COLLINA for the skilful construction of the equipments.

RIASSUNTO

Si descrive un esperimento con il quale è stato tentato di mettere in evidenza radiazione γ energetica proveniente da un particolare oggetto celeste (Cigno A) supposto da alcuni autori essere una collisione tra una galassia di materia ed una di anti-materia. L'esperimento ha dato risultato negativo, ed ha permesso di stabilire per il flusso γ un limite superiore di circa $5 \cdot 10^{-3}$ quanti/cm²s, valore di circa due ordini di grandezza inferiore a quello stimato da Morrison nell'ipotesi precedentemente menzionata.

The Renormalization Constants in Perturbation Theory.

J. G. TAYLOR

Department of Applied Mathematics and Theoretical Physics, Cambridge University

(ricevuto il 20 Maggio 1960)

Summary. — A simple theory of products of distributions is applied to the perturbation expansions of quantum field theories. This product requires that all divergent momentum integrals be replaced by finite but arbitrary constants. This product must be associative and commutative, and this can be satisfied by suitable choices of the arbitrary constants. In renormalizable theories any remaining arbitrariness only appears in the mass and charge, but will appear in other observables in non-renormalizable theories. These theories now become mathematically consistent but only practically useful for weak interactions.

1. — Introduction.

The renormalized perturbation solutions of the field equations of quantum electrodynamics have given very close agreement between theory and experiment, as has been shown, for example, for the Lamb shift and the anomalous magnetic moment of the electron. The chain linking the field equations and the experimental results has two especially weak links in it, namely the possible non-convergence of the perturbation expansions, and the appearance of infinite quantities in many of the terms of these expansions. We will not discuss the first weakness here, and we will assume that the perturbation expansions converge at least asymptotically. In this paper we will be concerned with the second weakness, and we will try to give a well-defined scheme of renormalization in perturbation theory with no infinite quantities occurring. Our discussion will apply to any field theory for which a perturbation expansion in powers of a small coupling constant has hopes of being a useful approximation to the complete solution.

To set the stage for our discussion let us briefly consider at what point the renormalization problem arises in the perturbation expansion of quantum electrodynamics. In this theory the S -matrix elements are evaluated by using Wick's ordering theorem which enables the T -products appearing in the expansion

$$(1) \quad S = \sum_{n=0}^{\infty} \frac{i^n}{n!} \int_{-\infty}^{+\infty} dx_1 \dots dx_n T [j_\mu(x_1) A^\mu(x_1) \dots j_\mu(x_n) A^\mu(x_n)]$$

to be replaced by normal ordered products (involving all annihilation operators on the right of all creation operators) with

$$(2) \quad N(A_1(x_1) \dots A_n(x_n)) = N(A_1(x_1) \dots A_n(x_n)) + \\ + \sum_{(1,2)} \widehat{A_1(x_1) A_2(x_2)} N(A_3(x_3) \dots A_n(x_n)) + \sum_{(1,2),(3,4)} \widehat{A_1 A_2 A_3 A_4} N(A_5(x_5) \dots A_n(x_n)) + \dots$$

The summation in the second term is over all pairs like (1, 2), in the third term over all pairs of pairs like (1, 2), (3, 4), and so on, whilst $\widehat{A_i A_j}$ is the usual contraction symbol. By this method we can immediately write down any matrix element of S by the usual graphical rules. However, the use of equation (2) gives rise to some poorly defined terms, for example the electron self-energy in lowest order is proportional to

$$\int dx_1 dx_2 N [\bar{\Psi}(1) \gamma_\mu S_F(x_1 - x_2) \gamma_\mu \Psi(2)] D_F(x_1 - x_2),$$

and involves the product $S_F(x) D_F(x)$. For small x^2 the functions Δ_F and D_F are singular like

$$\Delta_F(x) \sim \delta(x^2) + (i/\pi x^2),$$

$$D_F(x) \sim \delta(x^2) + (i/\pi x^2) - m^2 \varepsilon(x)/16\pi +$$

$$+ (m^2/8\pi^2) \left[\theta(x^2) \ln \frac{1}{2} m \sqrt{x^2} + i\theta(-x^2) \ln \frac{im}{2} \sqrt{-x^2} \right] + \text{less singular terms},$$

$$S_F(x) = (\gamma_\mu \partial_\mu - m) \Delta_F(x).$$

In order to define $S_F \cdot D_F$ it will be necessary to give meaning to the products $(\delta(x^2))^2$, $\delta(x^2)(1/x^2)$, $\delta(x^2)\delta(x_0)$, $\delta(x^2) \ln \sqrt{x^2}$, etc.

We may approach this problem in an obvious way by writing $[\delta(x)]^2 = \delta(0)\delta(x) = \infty\delta(x)$, but this is undefined since we do not wish to juggle

with such weighty things as infinite constants. It does not help to use approximating sequences, for example $\delta(x) = \lim_{n \rightarrow \infty} \exp[-nx^2] \sqrt{n/\pi}$, since $\int \exp[-2nx^2] \cdot (n/\pi) \varphi(x) dx \sim \sqrt{n} \varphi(0) \rightarrow \infty$ as $n \rightarrow \infty$ for any suitably well-behaved test function (x). So it seems necessary to investigate the problem of products of distributions more closely.

The methods that have been used up to now have been to avoid the singularities altogether which appear in the propagators, either by the regularization method of Pauli and Villars or by the use of a cut-off. These methods are *ad hoc* procedures for defining the products, and are certainly not contained in the basic equations of the theory. Further the values of the finite cut-offs are not given by the theory. This gives rise to an incomplete theory to which must be added a theory of the cut-offs. If the cut-off is allowed to become infinite then the results may not be defined, or may be different from the results obtained without a cut-off procedure. For these reasons, and also that we may understand something about non-renormalizable interactions we will try to develop a direct method for defining products of distributions.

The problem of defining a general product of distributions has been discussed at a deep level by KÖNIG ⁽¹⁾ in a series of papers. This definition has been applied to the products arising in quantum field theory by GÜTTINGER ⁽²⁾, and a similar definition has also been applied recently by BREMERMAN ⁽³⁾. It was claimed, though not fully proved, in each of these applications that the renormalization constants in each order of perturbation theory are finite but arbitrary. A simpler approach to the products of distributions will be developed here and applied to field theory. After an analysis of the problem of commutativity and associativity for the product we will reach the same conclusion about the renormalization constants, and also derive some results about non-renormalizable theories.

2. - Products of distributions.

Distributions ⁽⁴⁾ are defined in a linear fashion, and if S, T are distributions no meaning can necessarily be given to their product ST . We have already discussed the absence of δ^2 , for example. The most natural method to define the product ST is as follows ⁽²⁾. We denote by $D(T)$ the set of

⁽¹⁾ H. KÖNIG: *Math. Ann.*, **128**, 420 (1954).

⁽²⁾ W. GÜTTINGER: *Progr. Theor. Phys.*, **13**, 612 (1955).

⁽³⁾ H. J. BREMERMAN: O.N.R. Technical Report no. 8 (1959), Department of Mathematics, Berkeley.

⁽⁴⁾ L. SCHWARTZ: *Theorie des Distributions*, vol. 1 and 2 (Paris, 1951).

$\varphi \in \mathcal{D}^{(s)}$ with $T\varphi \in \mathcal{D}$. In other words $D(T)$ is the set of functions which vanish, together with enough derivatives, at points of singularity of T so that T is indefinitely differentiable at these points. We now define $\int (S \cdot T)(x) \varphi(x) dx = \int S(x) \cdot T\varphi(x) dx$, thus defining the product $S \cdot T$ as a linear functional on \mathcal{D} .

We will only consider cases where $S \cdot T$ reduces to a continuous function on $D(T)$, and so will be continuous in the topology induced on $D(T)$ by that on \mathcal{D} . We may then extend $S \cdot T$ from $D(T)$ so as to be a continuous linear functional on the whole of \mathcal{D} , and so a distribution. There will be many possible extensions, so there is an arbitrariness of the product.

As an example we consider the definition of δ^2 . Evidently $\delta\varphi \in \mathcal{D}$ if $\varphi(0) = 0$, so that $\delta\varphi = 0$ and $D(\delta) = [\varphi : \varphi(0) = 0]$ ⁽⁶⁾. Hence if $\Psi \in D(\delta)$ then $\int \delta^2 \cdot \Psi = \int \delta \cdot \delta\Psi = 0$. To extend δ^2 to the whole of \mathcal{D} we choose a fixed $\alpha \in \mathcal{D}$ with $\alpha(0) = 1$ and write, for any $\varphi \in \mathcal{D}$, $\varphi = \varphi(0)\alpha + \Psi$, $\Psi \in D(\delta)$. Then $\int \delta^2 \cdot \varphi = (\int \delta^2 \cdot \alpha)\varphi(0) = c\varphi(0)$, where c is the constant $\int \delta^2 \cdot \alpha$ which may be chosen arbitrarily. Thus $\delta^2 = c\delta$, and the arbitrariness appears in the constant c . Similarly we may show that $\delta'\delta = c'\delta$, where c' is another arbitrary constant.

When we consider $\delta\delta'$, $D(\delta') = [\varphi : \varphi(0) = \varphi'(0) = 0]$, and for $\Psi \in D(\delta')$ we have $\int \delta\delta' \cdot \Psi = 0$. We extend the definition of $\delta\delta'$ from $D(\delta')$ to \mathcal{D} by choosing fixed $\alpha, \beta \in \mathcal{D}$ with $\alpha(0) = \beta'(0) = 1$, $\alpha'(0) = \beta(0) = 0$, so that any φ can be written $\varphi = \varphi(0)\alpha + \varphi'(0)\beta + \Psi$, $\Psi \in D(\delta')$.

Hence $\int \delta\delta' \cdot \varphi = (\int \delta\delta' \cdot \alpha)\varphi(0) + (\int \delta\delta' \cdot \beta)\varphi'(0)$, so $\delta\delta' = c_1\delta + c_2\delta'$, where $c_1 = \int \delta\delta' \cdot \alpha$ and $c_2 = \int \delta\delta' \cdot \beta$ are arbitrary constants. Similar discussions may be given for the products of higher derivatives of δ -functions.

In order to reduce some of the ambiguity of the product we impose the condition that differentiation is distributive: $(ST)' = S'T + ST'$. Then $(\delta\delta') = \delta'\delta + \delta\delta'$, so that the constants appearing in our discussion of these products are related by $c = c_2$, $c_1 + c' = 0$. So far we have not discussed the problem of associativity or commutativity of the product.

In general we cannot require an associative product, as we see from the simple counter-example $0 = x^{-1}(x\delta) \neq (x^{-1}x)\delta = \delta$. We may require commutativity, but then we see that for the examples treated above $c_2 = 0$, $c_1 = c'$, so that $c_1 = c_2 = c' = 0$ and the products δ^2 , $\delta\delta'$, $\delta'\delta$ all vanish. This is true also for products of higher derivatives, so we see that commutativity removes all arbitrariness in the products of derivatives of δ -functions, though at the same time it removes the products.

⁽⁵⁾ We use the notation of ref. (4) throughout. \mathcal{D} denotes the set of indefinitely differentiable functions vanishing outside a bounded set, and \mathcal{D}' denotes the set of distributions of ref. (4). $a \in A$ denotes a belongs to the set A .

⁽⁶⁾ The set of x for which $P(x)$ is true is denoted by $[x : P(x)]$.

Similarly if we require $\delta_{(a)}\delta_{(b)} = \delta_{(b)}\delta_{(a)}$ and since $\delta_{(a)}\delta_{(b)} = e_b\delta_{(b)}$, $\delta_{(b)}\delta_{(a)} = e_a\delta_{(a)}$ then commutativity implies that arbitrary constants e_a , e_b both vanish. Thus again products of derivatives of δ -functions at different points commute if and only if they vanish.

So far we have introduced a product of any pair of distributions which satisfy very general conditions. The possible lack of associativity and commutativity and the arbitrariness arising in the product are unpleasant features of such a general product. We may obtain commutativity by reducing some of the arbitrariness, though this may not always be possible.

3. - Products in quantum electrodynamics.

If we do not have commutativity or associativity of the product then the usual renormalization procedure of quantum electrodynamics cannot be used and physical expressions involve other constants than masses and charges of particles. It is evidently necessary to investigate if we may define a commutative and associative product for the products of the singular functions entering in field theory. Indeed, since the T -product is independent of the order of the operators it contains, the products of the singular functions S_F and D_F must be commutative and associative wherever they occur in the normal ordering expansion of the T -product. We will see that these conditions will impose restrictions on the arbitrary coefficients arising in the products which can be satisfied.

As the simplest example let us consider the product $S_F D_F$ arising in the lowest order electron self-energy, given in momentum space by

$$(1) \quad \Sigma(p) = \int \frac{d^4k}{k^2} \frac{i\gamma(p-k) - m}{(p-k)^2 - m^2}.$$

We regard $\Sigma(p)$ as a poorly defined distribution. It is defined for the class $D(\Sigma)$ of functions for which $\int \varphi dp = \int p_\mu \varphi dp = 0$ ($\mu = 1, 2, 3, 4$). If we expand Σ formally about $p = 0$, $\Sigma = A + B_\mu p_\mu + c(p)$, then

$$\int \Sigma \varphi dp = \int c(p) \varphi dp$$

which is finite since $c(p) = p_\mu p_\nu (\partial^2 \Sigma / \partial p_\mu \partial p_\nu)_{p=\xi}$, which is a finite function since it involves two extra powers of k in the denominator of the integrand defining Σ . We extend the definition of Σ to all $\Phi \in \mathcal{D}$ by choosing fixed α, β_μ with $\int \alpha dp = 1$, $\int p_\mu \beta dp = \delta_{\mu\nu}$, $\int p_\mu \alpha dp = \int \beta_\mu dp = 0$. Then any $\varphi \in \mathcal{D}$ may be written

$$\varphi = \left(\int \varphi dp \right) \alpha + \left(\int p_\mu \varphi dp \right) \beta_\mu + \Psi, \quad \text{with } \Psi \in D(\Sigma),$$

and

$$\int \Sigma \varphi \, dp = \left(\int \Sigma x \, dp \right) \int \varphi \, dp + \left(\int \Sigma \beta_\mu \, dp \right) \int p_\mu \varphi \, dp + \int c(p) \Psi \, dp$$

or

$$\Sigma = a + b_\mu p_\mu + c(p),$$

where $a = \int \Sigma x \, dp - \int c x \, dp$, $b_\mu = \int \Sigma \beta_\mu \, dp - \int c \beta_\mu \, dp$, are arbitrary constants, and $c(p)$ is a finite continuous function. In the usual way we can write

$$\Sigma = a + b i \gamma p + m + \Sigma_f (i \gamma p + m)^2,$$

where a, b are finite and arbitrary constants and Σ_f is a unique and finite function of p .

If we consider the difference between SD and DS we note that the «finite parts» will be the same in both cases. To consider the conditions on the arbitrary parts so that SD and DS are equal we only need consider this equality when the Fourier transforms of S and D are $(ik\gamma - m)/k^2$ and $1/k^2$, since additional terms give finite contributions. We consider the arbitrary constants a, b as replaced formally by their expressions in terms of $\int d^4k (k^2)^{-2}$. We see that for these values of the arbitrary constants the «infinite parts» of SD and DS are equal, as is verified from the equation

$$\int [i \gamma_\mu (p_\mu - k_\mu) + m] / (p - k)^2 k^2 = \int (i_\mu k_\mu + m) / (p - k^2) k^2$$

for the finite parts of each side are identical, whilst the infinite part of the left hand side is $(\frac{1}{2} i \gamma_\mu p_\mu + m) \int (k^2)^{-2} d^4k$, as is that of the right-hand side. What we have done for $\Sigma(p)$ is to replace the infinite part $\int (k^2)^{-2} d^4k$ by an arbitrary constant, and this ensures commutativity of the product SD , or in momentum space of the convolution product of the Fourier transforms of S and D . In other places where the formally infinite constants $\int (k^2)^{-n} d^4k$ ($n = 1, 2$) arise we may replace them by arbitrary constants. This ensures that the products arising in quantum electrodynamics are both associative and commutative, for this replacement is formally equivalent to the introduction of a cut-off in the momentum integrals. We emphasize that this equivalence between a cut-off theory and the product theory defined here is not to be taken seriously. In particular there is no physical reason for choosing one value of the arbitrary constants rather than another, whilst a cut-off value much below the mass of the heaviest particle taking part is physically unreasonable. Further the product does not involve any change in the local nature of the interaction.

In this manner we have obtained a product theory of distributions which never allows infinite quantities to enter in the perturbation calculations for quantum electrodynamics. Since this product contains arbitrary constants chosen so that the product is associative and commutative and also so that, for example, gauge invariance is satisfied, then the renormalization programme shows that these arbitrary constants only appear as alterations to the mass and charge of the electron. In any case we take these quantities from experiment, so this ambiguity is not troublesome.

In the case of other field theories which have perturbation expansions which are renormalizable by the usual methods we see in a similar manner that our product of distributions ensures that the renormalization prescription is applied only to finite quantities. Also any ambiguity arising in the product is removed completely by renormalization, except that masses and coupling constants are altered in an arbitrary fashion, and may only be determined directly from experiment.

4. — Products in non-renormalizable theories.

So far nothing new has been obtained by our product. This is not the case for non-renormalizable theories, which we will consider briefly now. For any interaction involving any number of fields and derivatives of them the convolution products arising in the perturbation expansion of the S -matrix can be made well-defined distributions in an exactly similar manner to that used for $\Sigma(p)$ in the previous section. Furthermore these convolution products may be made associative and commutative by choosing the arbitrary constants which arise in them to satisfy the same algebraic relations which they would if they were regarded as being defined in terms of the infinite quantities $\int (k^2)^n d^4k$ ($n \geq -2$). There will be an infinite number of such arbitrary constants now, and these cannot be eliminated from observable quantities by renormalization. Thus all these constants will have to be determined from experiment.

We see that the product definition gives a mathematically consistent method of using non-renormalizable theories. It will not be very practical to use this method for strong interactions, not only because of the possible lack of convergence of the perturbation expansion but also because of the large number of parameters which will arise in matrix elements. It will be in weak interactions, where only the first or second terms of the perturbation expansions are important, that the matrix elements will involve only one or two arbitrary parameters to be determined from experiment. In such cases, for example the vector intermediate meson in β -decay, the theory is both mathematically consistent and practically useful.

5. - Discussion.

There are certain models for which closed form solutions may be written down (^{7,8}). In these solutions no arbitrary constants appear, and it may be questioned whether or not the arbitrariness arising in our results is due to the perturbation expansions, and would not occur by a closed form solution. In the case of the Lee model (⁷) the results obtained by taking no cut-off involve infinite quantities (which, of course, never enter observable quantities) and so suffer from not being properly defined. Their proper definition gives rise to arbitrary but finite renormalization constants.

For the derivative coupling theories (⁸) which give a propagator $\exp [ig^2 \Delta_F(x)]$ we see that in order to make this quantity well-defined we have to consider $(\Delta_F(x))^n$, and this will certainly involve arbitrary constants. In other words these closed form solutions hide the arbitrariness which will result when they are evaluated explicitly.

We conclude that the arbitrariness need not arise from the use of a perturbation expansion, and that even in closed form solutions this arbitrariness persists.

(⁷) T. D. LEE: *Phys. Rev.*, **95**, 1329 (1954).

(⁸) R. ARNOWITT and S. DESER: *Phys. Rev.*, **100**, 349 (1955); L. N. COOPER: *Phys. Rev.*, **100**, 362 (1955).

RIASSUNTO (*)

Una semplice teoria dei prodotti di distribuzione è applicata agli sviluppi di perturbazioni delle teorie quantistiche del campo. Questo prodotto richiede che tutti gli integrali divergenti della quantità di moto siano sostituiti da costanti finite ma arbitrarie. Questo prodotto deve godere delle proprietà associativa e commutativa, e questo può essere ottenuto con una opportuna scelta delle costanti arbitrarie. Nelle teorie rinormalizzabili qualsiasi arbitrarietà rimanente compare solo nella massa e nella carica, ma apparirà in altre osservabili nelle teorie non rinormalizzabili. Queste ultime ora divengono matematicamente consistenti ma praticamente utili solo nelle interazioni deboli.

(*) Traduzione a cura della Redazione.

On the Quantum Conditions in Field Theory - IV.

R. S. LIOTTA (*)

Department of Physics, Columbia University - New York, N. Y.

(ricevuto il 25 Maggio 1960)

Summary.— We apply the covariant functional formalism to the Feynman-Gell-Mann spinorial equation. We extend the first order approximation of the W.K.B. method to field theory, giving the corresponding quantum conditions. Applying these results to free fields, we find a relationship between various quantum numbers in field theory. For coupled fields we find indications for a further treatment of the problem.

1. - Introduction.

In a previous series of articles ⁽¹⁾ we developed a covariant functional formalism for free and interacting fields placing the emphasis on the properties of internal coherence of the theory. Thus one obtains in effect, by means of the zero order approximation of the W.K.B. method which is extended to field theory and applied to the equation of motion,

$$(1) \quad \mathcal{H}\bar{\Psi} = i\hbar c \frac{\delta \bar{\Psi}}{\delta \sigma(x)},$$

in the Schrödinger representation, as has been shown in the aforementioned articles.

(*) On leave of absence from Istituto di Fisica dell'Università, Roma and Istituto Nazionale di Fisica Nucleare, Sezione di Roma.

⁽¹⁾ R. S. LIOTTA: *Nuovo Cimento*, **3**, 438 (1956); **8**, 798 (1958); **13**, 921 (1959); referred to respectively as Parts I, II, III; and **14**, 443 (1959).

On the other hand, it is well known that the first order approximation of this method, corresponds in the usual Schrödinger theory, to the solution of the problem of eigenvalues of the equation of motion for closed problems, and justifies the quantum condition empirically postulated by BOHR ⁽²⁾, SOMMERFELD ⁽³⁾, WILSON ⁽⁴⁾ and ISHIWARA ⁽⁵⁾ (B.S.W.I.) from the point of view of wave mechanics.

This first order approximation allows for an ample physical interpretation of the theory and gives a sufficient number of results comparable to experiments.

Extending this approximation to field theory, we shall see that it is possible to obtain results subject to immediate experimental interpretation. However, this method reveals itself to be analytically less powerful than the analogous method applied to systems of a finite number of degrees of freedom, in so much as this method gives the eigenvalue spectra of $\int T(x) d\sigma$ for stationary problems, but gives no information on the value of $\hat{T}(x)$ for each point of a space-like surface; $T(x)$ being the generic eigenvalue of the operator \mathcal{H} in (1) and $\int T(x) d\sigma$ being the integral on a generic space-like surface.

We shall apply this method to various types of free and interacting fields of interest in physics, recalling that this approximation does not give way to the inconveniences of perturbation theory since it is independent of coupling constants.

First however, it will be necessary to give completely uniform analytical criteria both for the boson and fermion fields, in order to overcome some of the mathematical complications which may be verified using the formalism developed in Part III for fermion fields and based on Dirac's equation.

It is useful to establish that these complications do not present themselves till we consider the problems in which the Dirac field is assigned, while other fields, for example the electromagnetic field, may vary determining the evolution of the system as we shall see below.

Section 2 will be therefore, an extension of methods discussed in the preceding articles for the case of free and coupled fermion fields, satisfying a second order equation as proposed by FEYNMAN and GELL-MANN ⁽⁶⁾ (F.G-M.).

Section 3 will attempt a separation of the equation (1) into two parts. One part being «time dependent» and the other «time independent». The conditions of B.S.W.I. both for separable and non-separable systems will be elaborated for field theory. This will be done following criteria recently per-

⁽²⁾ N. BOHR: *Phil. Mag.*, **26**, 1 (1913).

⁽³⁾ W. WILSON: *Phil. Mag.*, **28**, 795 (1915).

⁽⁴⁾ ISHIWARA: *Tokyo Math. Phys. Proc.*, **8**, 106 (1915).

⁽⁵⁾ A. SOMMERFELD: *Ann. der Phys.*, **51**, 1 (1916).

⁽⁶⁾ R. P. FEYNMAN and M. GELL-MANN: *Phys. Rev.*, **109**, 193 (1958).

feeted by KELLER⁽⁷⁾ for systems with a finite number of degrees of freedom. We shall see how quantum conditions correspond to the solution of the problem of eigenvalues for the equation of motion within certain limits.

Section 4 will discuss the physical meaning of the quantity $\int T(x) dx$ and will give the mass spectra for free fields as eigenstates of this integral. We will also try to give the same significance of mass for some coupled fields, but it will be necessary to add an *ad hoc* hypothesis to the V region of a space-like surface where we extend the previous integral.

The verification of this adjunct hypothesis may be determined by the complete solution of the integro-differential equation of motion. This will be explained in a later work in which we will also give more detailed considerations to functional space. In this paper we will use some of the properties of this space⁽⁸⁾.

It is necessary to establish here that in the following sections we shall make use of units of measure different from those adopted in Parts I, II, III, so as to establish more clearly the numerical results.

Furthermore, in the differential operators of second quantization we will use the imaginary unit i explicitly so that the eigenstates of these operators will have the same space-like or time-like character as the operators used at the outset.

2. - Covariant functional formalism for spinor fields, satisfying the Feynman-Gell-Mann equation, interacting with the electromagnetic field.

For the following developments it is necessary to note here briefly some of the considerations which led to the Feynman-Gell-Mann equation.

We start with the Dirac equation for electrons which interact with the electromagnetic field:

$$(2) \quad \left\{ \gamma_\mu \left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c} A_\mu \right) + k_0 \right\} \psi = 0,$$

$$(3) \quad \left\{ \gamma_\mu^x \left(\frac{\partial}{\partial x_\mu} + \frac{ie}{\hbar c} A_\mu \right) - k_0 \right\} \bar{\psi} = 0,$$

⁽⁷⁾ J. B. KELLER: *Ann. Phys.*, **4**, 180 (1958).

⁽⁸⁾ L. FANTAPPIÈ: *I funzionali analitici*, in *Mem. Acc. Naz. Linc.* serie VI, **3**, fasc. XI (1930).

where $\gamma_\mu \gamma_\nu + \gamma_\nu \gamma_\mu = 2\delta_{\mu\nu}$; γ_μ^T is the transposed matrix of γ_μ , $k_0 = \frac{m_0 c}{\hbar}$.

Placing:

$$(4) \quad \psi = \frac{1}{k_0} \left\{ \gamma_\mu \left(\frac{\partial}{\partial x_\mu} - \frac{ie}{\hbar c} A_\mu \right) - k_0 \right\} \varphi,$$

$$(5) \quad \bar{\psi} = \frac{1}{k_0} \left\{ \gamma_\mu^T \left(\frac{\partial}{\partial x_\mu} + \frac{ie}{\hbar c} A_\mu \right) + k_0 \right\} \bar{\varphi},$$

we obtain the second order equations:

$$(6) \quad \frac{\partial^2 \varphi}{\partial x_\mu^2} - \frac{2ie}{\hbar c} \frac{\partial \varphi}{\partial x_\mu} A_\mu + \frac{e}{2\hbar c} \sigma_{\mu\nu} F_{\mu\nu} \varphi - \frac{e^2}{\hbar^2 c^2} A_\mu^2 \varphi - k_0^2 \varphi = 0,$$

$$(7) \quad \frac{\partial^2 \bar{\varphi}}{\partial x_\mu^2} + \frac{2ie}{\hbar c} \frac{\partial \bar{\varphi}}{\partial x_\mu} A_\mu + \frac{e}{2\hbar c} \sigma_{\mu\nu}^T F_{\mu\nu} \bar{\varphi} - \frac{e^2}{\hbar^2 c^2} A_\mu^2 \bar{\varphi} - k_0^2 \bar{\varphi} = 0,$$

where

$$\sigma_{\mu\nu} = -\frac{i}{2} (\gamma_\mu \gamma_\nu - \gamma_\nu \gamma_\mu), \quad F_{\mu\nu} = \frac{\partial A_\nu}{\partial x_\mu} - \frac{\partial A_\mu}{\partial x_\nu},$$

and $\sigma_{\mu\nu}^T$ is the transposed matrix of $\sigma_{\mu\nu}$.

The solutions of these equations may be placed in a one to one correspondence with the solutions of equations (2), (3) imposing the conditions

$$(8) \quad \gamma_5 \varphi = \varphi, \quad \gamma_5 \bar{\varphi} = \bar{\varphi}.$$

Then for each ψ , $\bar{\psi}$ there is respectively only one φ , $\bar{\varphi}$ given by

$$(9) \quad \varphi = -\frac{1}{2}(1 + \gamma_5)\psi, \quad \bar{\varphi} = -\frac{1}{2}(1 + \gamma_5)\bar{\psi}$$

and spinorial functions φ , $\bar{\varphi}$ result in only two independent components given by the first two components. These preliminary considerations are sufficient for further developments and thus naturally for any discussion of their significance we refer the reader to the work of FEYNMAN and GELL-MANN.

We may assume the function:

$$(10) \quad L = \frac{\hbar^2}{m_0} \left\{ - \left(\frac{\partial \bar{\varphi}}{\partial x_\mu} + \frac{ie}{\hbar c} A_\mu \bar{\varphi} \right) \left(\frac{\partial \varphi}{\partial x_\mu} - \frac{ie}{\hbar c} A_\mu \varphi \right) + \frac{e}{2\hbar c} \bar{\varphi} \sigma_{\mu\nu} F_{\mu\nu} \varphi - k_0 \bar{\varphi} \varphi \right\} -$$

$$- \frac{1}{2} \frac{\partial A_\mu}{\partial x_\nu} \frac{\partial A_\mu}{\partial x_\nu},$$

as a Lagrangian function for the interaction of electromagnetic and spinor fields; it gives the correct equation of motion.

By the Lorentz condition we have:

$$(11) \quad \frac{e}{2\hbar c} \sigma_{\mu\nu} F_{\mu\nu} = \frac{ie}{\hbar c} \gamma_\mu \gamma_\nu \frac{\partial A_\mu}{\partial x_\nu}; \quad \frac{e}{2\hbar c} \sigma_{\mu\nu}^* F_{\mu\nu} = \frac{ie}{\hbar c} \gamma_\mu^* \gamma_\nu^* \frac{\partial A_\mu}{\partial x_\nu}.$$

So that for the conjugate momenta (see Part I) we obtain:

$$(12) \quad P_{\mu\nu} = \frac{4}{c} \frac{\partial L}{\partial (\partial A_\mu / \partial x_\nu)} = -\frac{4}{c} \left(\frac{\partial A_\mu}{\partial x_\nu} - \frac{ie}{k_0} \bar{\varphi} \gamma_\mu \gamma_\nu \varphi \right),$$

$$(13) \quad \chi_\mu = \frac{4}{c} \frac{\partial L}{\partial (\partial \bar{\varphi} / \partial x_\mu)} = -\frac{4}{c} \left(\frac{\partial \bar{\varphi}}{\partial x_\mu} + \frac{ie}{\hbar c} A_\mu \bar{\varphi} \right) \frac{\hbar^2}{m_0},$$

$$(14) \quad \bar{\chi}_\mu = \frac{4}{c} \frac{\partial L}{\partial (\partial \varphi / \partial x_\mu)} = -\frac{4}{c} \left(\frac{\partial \varphi}{\partial x_\mu} - \frac{ie}{\hbar c} A_\mu \varphi \right) \frac{\hbar^2}{m_0},$$

(for $\mu = \nu$ the last term in (12) is zero) and for the trace of canonical tensor:

$$(15) \quad H = -\frac{e^2}{8} P_{\mu\nu} P_{\mu\nu} + \frac{ie}{\hbar} (\chi_\mu \varphi - \bar{\chi}_\mu \bar{\varphi}) A_\mu + \frac{ie c}{k_0} P_{\mu\nu} \bar{\varphi} \gamma_\mu \gamma_\nu \varphi - \\ - \frac{k_0 c}{4\hbar} \bar{\chi}_\mu \chi_\mu + 4m_0 c^3 \bar{\varphi} \varphi + \frac{2e^2}{k_0^2} (\bar{\varphi} \gamma_\mu \gamma_\nu \varphi)^2.$$

It is easy to verify that the equations of motion are given by the generalized hamiltonian system (Part I, eq. (9)) using the hamiltonian (15). It is not necessary to use any precautions for the orders, as previously occurred using the hamiltonian given in Part III, eq. (46).

In order to construct equation (1) it is sufficient to replace the conjugate momenta $P_{\mu\nu}$, $\bar{\chi}_\nu$, χ_ν , with the corresponding operators:

$$(16) \quad \begin{cases} \mathcal{P}_{\mu\nu}(x) = -i\hbar n_\nu \frac{\delta}{\delta A_\mu(x)}, \\ (\bar{\chi}_\nu(x))_\alpha = -i\hbar n_\nu \frac{\delta}{\delta \bar{\varphi}_\alpha(x)}; \quad (\chi_\nu(x))_\alpha = -i\hbar n_\nu \frac{\delta}{\delta \varphi_\alpha(x)}, \end{cases}$$

where α is the spinorial index, and by considering the Lagrangian co-ordinate a multiplicative operator. The state vector is expressed as a complex functional of the type

$$\bar{\Psi}(\varphi_\alpha, \bar{\varphi}_\beta, A_\mu, \sigma) = \bar{B}(\varphi_\alpha, \bar{\varphi}_\beta, A_\mu, \sigma) \exp \left[\frac{i}{\hbar} \bar{V}(\varphi_\alpha, \bar{\varphi}_\beta, A_\mu, \sigma) \right],$$

where α, β are spinorial indices and \bar{B} and \bar{V} are real functionals of the form

$$\bar{B}(\varphi_\alpha, \bar{\varphi}_\beta, A_\mu, \sigma) = \int_{\sigma} B(\varphi_\alpha, \bar{\varphi}_\beta, A_\mu, x) d\sigma,$$

and they are obviously c numbers.

For the electromagnetic field we have the commutators (see Part II, eq. (19))

$$(17) \quad [\mathcal{P}_{\mu\nu}(x), \mathcal{A}_\lambda(x')] = -i\hbar \delta_{\lambda\mu} \frac{\partial}{\partial x_\nu} D(x - x').$$

For the Feynman-Gell-Mann field (the reader is referred to Parts II and III) let us consider the isolated spinorial field. If $\bar{\gamma}_\lambda(x)$ is a spinor function belonging to the solutions in equation (7), without the electromagnetic field, such that $\bar{\Psi}(\bar{\varphi}_\alpha, \varphi_\beta, \sigma) \bar{\gamma}_\lambda(x) \varphi_\lambda(x)$ is a c number, we have

$$\begin{aligned} \int_{\sigma} n_\nu(x) \frac{\delta}{\delta \varphi_\mu(x)} [\bar{\Psi}(\bar{\varphi}_\alpha, \varphi_\beta, \sigma) \bar{\gamma}_\lambda(x) \varphi_\lambda(x')] d\sigma(x) = \\ = - \int_{\sigma} \varphi_\lambda(x') n_\nu(x) \frac{\delta}{\delta \varphi_\mu(x)} [\bar{\Psi}(\varphi_\alpha, \varphi_\beta, \sigma) \gamma_\lambda(x)] d\sigma(x) + \delta_{\mu\lambda} \bar{\Psi}(\bar{\varphi}_\alpha, \varphi_\beta, \sigma) \bar{\gamma}_\lambda(x'), \end{aligned}$$

thus

$$n_\nu(x) \frac{\delta}{\delta \varphi_\mu(x)} \{ \bar{\Psi}(\bar{\varphi}_\alpha, \varphi_\beta, \sigma) \varphi_\lambda(x') \} + \varphi_\lambda(x') n_\nu(x) \frac{\delta}{\delta \varphi_\mu(x)} \bar{\Psi}(\bar{\varphi}_\alpha, \varphi_\beta, \sigma) = \delta_{\lambda\mu} \delta_\nu(x - x') \bar{\Psi},$$

where

$$\delta_\nu(x - x') = \frac{\partial}{\partial x_\nu} \Delta(x - x'),$$

and Δ is the Pauli-Jordan function for the field under consideration.

Therefore we can write

$$(18) \quad \{ (\chi_\nu(x))_\mu, \varphi_\lambda(x') \}_+ = -i\hbar \delta_{\lambda\mu} \frac{\partial}{\partial x_\nu} \Delta(x - x'),$$

and the analogous for $(\bar{\chi}_\nu)_\mu, \bar{\varphi}_\lambda$ for the canonical conjugate operators, while other operators follow the ordinary rules of anticommutation⁽⁹⁾.

It follows that all the considerations expressed for free and interacting fields satisfying second order equations, can be extended to this case.

(9) J. SCHWINGER: *Phys. Rev.*, **74**, 1439 (1948).

In particular (1) may be broken down into real and imaginary parts:

$$(19) \quad \text{Re) } \frac{c^2}{4} \left\{ \frac{1}{\varepsilon} \left(\frac{\delta \bar{V}}{\delta A_\mu} \right)^2 \bar{B} - \hbar^2 \frac{\delta^2 \bar{B}}{\delta A_\mu^2} + \frac{k_0}{\hbar c} \frac{\delta \bar{V}}{\delta \bar{\varphi}} \frac{\delta \bar{V}}{\delta \varphi} \bar{B} + \frac{k_0}{\hbar c} \hbar^2 \frac{\delta^2 \bar{B}}{\delta \bar{\varphi} \delta \varphi} \right\} +$$

$$+ \frac{ie c}{k_0} n_\nu \bar{\varphi} \gamma_\mu \gamma_\nu \varphi \frac{\delta \bar{V}}{\delta A_\mu} \bar{B}' + \frac{2e^2}{\hbar^2} (\bar{\varphi} \gamma_\mu \gamma_\nu \varphi)^2 \bar{B} + 4m_0 c^2 \bar{\varphi} \varphi \bar{B} +$$

$$+ \frac{ie}{\hbar} \left(\varphi \frac{\delta \bar{V}}{\delta \bar{\varphi}} - \bar{\varphi} \frac{\delta \bar{V}}{\delta \varphi} \right) n_\nu A_\nu \bar{B} = -e \frac{\delta \bar{V}}{\delta \sigma(x)} \bar{B},$$

$$(20) \quad \text{Im) } -\frac{1}{4} \left\{ \frac{\delta}{\delta A_\mu} \left[\left(e \frac{\delta \bar{V}}{\delta A_\mu} + \frac{4ie}{k_0} n_\nu \bar{\varphi} \gamma_\mu \gamma_\nu \varphi \right) \bar{B}^2 \right] + \frac{\delta}{\delta \varphi} \left[\left(e \frac{\delta \bar{V}}{\delta \bar{\varphi}} + \frac{4ie}{k_0} n_\nu A_\nu \varphi \right) \bar{B}^2 \frac{k_0}{\hbar c} \right] + \right.$$

$$\left. + \frac{\delta}{\delta \bar{\varphi}} \left[\left(e \frac{\delta \bar{V}}{\delta \varphi} - \frac{4ie}{k_0} n_\nu A_\nu \bar{\varphi} \right) \bar{B}^2 \frac{k_0}{\hbar c} \right] \right\} = \frac{\delta \bar{B}^2}{\delta \sigma(x)},$$

and it may be easily verified that the Lorentz condition is compatible with this scheme (see Part III, eq. (52), (53)).

It is useful to consider here the case in which the density of probability \bar{B}^2 is independent of the space-like surface, *i.e.*, $\delta \bar{B}^2 / \delta \sigma(x) = 0$. Let us consider a trajectory tube in functional space of A_μ , $\bar{\varphi}$, φ ; applying Gauss' theorem in this space, and considering

$$(21) \quad \begin{cases} U_{A_\mu} = -\frac{1}{4} \left(e \frac{\delta \bar{V}}{\delta A_\mu} + \frac{4ie}{k_0} n_\nu \bar{\varphi} \gamma_\mu \gamma_\nu \varphi \right), \\ U_\varphi = -\frac{1}{4} \frac{k_0}{\hbar c} \left(e \frac{\delta \bar{V}}{\delta \bar{\varphi}} + \frac{4ie}{k_0} n_\nu A_\nu \varphi \right); \\ U_{\bar{\varphi}} = -\frac{1}{4} \frac{k_0}{\hbar c} \left(e \frac{\delta \bar{V}}{\delta \varphi} - \frac{4ie}{k_0} n_\nu A_\nu \bar{\varphi} \right), \end{cases}$$

as functional components of the velocity $U(A_\mu, \bar{\varphi}, \varphi)$, the «current density» of the field $\bar{\Psi}(A_\mu, \bar{\varphi}, \varphi, \sigma)$ is given by

$$\bar{J}(A_\mu, \bar{\varphi}, \varphi) = \bar{B}^2 U(A_\mu, \bar{\varphi}, \varphi)$$

and we can write (20) in the form

$$(20') \quad \bar{J}(A_\mu, \bar{\varphi}, \varphi) = \bar{J}_0 \frac{d\Sigma_0}{d\Sigma},$$

where $d\Sigma$ is the «area» of the normal section of the trajectory tube around the point A_μ , $\bar{\varphi}$, φ , which is at one end of the tube, and the index 0 is relative to the corresponding functions at the other end of the tube.

3. - Stationary form of the equation of motion.

From considerations developed in previous sections of this paper and in Parts I, II, III, it results that the equation of motion of first quantization both for free and coupled fields can be written in the generalized hamiltonian form

$$(22) \quad e \frac{\partial q_k(x)}{\partial x_\nu} = \frac{\partial H}{\partial p_{kv}(x)}; \quad e \frac{\partial p_{kv}(x)}{\partial x_\nu} = - \frac{\partial H}{\partial q_k(x)},$$

or by the Jacobi generalized equation

$$(23) \quad H \left(q_k(x) \frac{\partial V_\nu(q_k(x), x)}{\partial q_k(x)} \right) + e \frac{\partial V_\nu(q_k(x), x)}{\partial x_\nu} = 0,$$

where $H(q_k(x), p_{kv}(x))$ is the trace of the canonical tensor and $q_k(x)$, $p_{kv}(x)$ are respectively the Lagrangian co-ordinates, functions of a generic point of space-time, and the conjugate momenta with respect to the four directions of the space-time. We will use the Latin indices for the components of the Lagrangian co-ordinates, which can be either of tensorial or spinorial character and we will use Greek indices for the tensorial components. The properties of an unknown function V_ν are defined in Parts I, II, III; it is sufficient here to remember that the tensor character of V_ν is such that $V_\nu(q_k, x) = n_\nu V(q_k, x)$, where V is an invariant function and $n_\nu(x)$ is the normal unit vector to the space-like surface in the point x . The function H is naturally an invariant.

The equation of motion of second quantization is (1), where the operator \mathcal{H} is a function of the point x , on σ , and is given by

$$(24) \quad \mathcal{H} = \mathcal{H} \left(q_k(x), -i\hbar n_\nu(x) \frac{\delta}{\delta q_k(x)} \right),$$

and the state vector is a complex functional of q_k and of the space-like surface:

$$(25) \quad \bar{\Psi}(q_k, \sigma) = \int_{\sigma} \Psi(q_k(x), x) d\sigma.$$

To find stationary solutions of (1) we suppose that the state vector (25) can be written in the form

$$(26) \quad \bar{\Psi}(q_k, \sigma) = \bar{\Phi}(q_k) \bar{R}(\sigma),$$

where $\bar{\Phi}(q_k)$ is a functional that does not depend explicitly from the space-like surface σ , while $\bar{R}(\sigma)$ depends only on the surface σ . Since the operator \mathcal{H} ope-

rates only on the q_k , we obtain from (1), as in the ordinary Schrödinger theory, two equations respectively, one «time-independent» and the other «time-dependent»:

$$(27) \quad \mathcal{H} \bar{\Phi}(q_k) = T(x) \bar{\Phi}(q_k),$$

$$(28) \quad -\frac{i}{\hbar c} T(x) d\omega = \frac{\delta \bar{R}(\sigma)}{R(\sigma)},$$

where $T(x)$ is a classical function of the point x of the space-like surface σ_0 in which equation (27) is defined and $d\omega$ is the element of volume between σ_0 and σ in space-time. The integral of (28) is given by

$$(29) \quad \bar{R}(\sigma) = R_0 \exp \left[-\frac{i}{\hbar c} \int_{\sigma_0}^{\sigma} T(x) d\omega \right].$$

If we introduce the parameter τ which defines the family of space-like surfaces with which we study the motion, the functional of state (25) can be written introducing the amplitude and phase (see Part II), in the form

$$(30) \quad \bar{\Psi}(q_k, \sigma) = \bar{B}(q_k) \exp \left[\frac{i}{\hbar} \bar{V}(q_k, \sigma) \right],$$

where

$$(31) \quad \bar{V}(q_k, \sigma) = \int_{\sigma} V_{\nu}(q_k(x), x) d\sigma_{\nu} = \int_{\sigma} n_{\nu} \left(S(q_k(x)) - \int_{\tau_0}^{\tau} T(x) d\tau \right) d\sigma_{\nu}.$$

We have conglobated the constant R_0 in $\bar{B}(q_k)$, $d\sigma_{\nu} = n_{\nu} d\sigma_{\nu}$, and $S(q_k)$ is a function of the q_k only. In fact we obtain directly

$$\frac{1}{c} \int_{\sigma_0}^{\sigma} T(x) d\omega = \int_{\tau_0}^{\tau} d\tau \int_{\sigma} T(x) d\sigma,$$

and if we suppose

$$(32) \quad \int_{\sigma} T(x) d\sigma = M,$$

with M constant with respect to τ , equation (31) follows.

In as much as the equation of motion (1) becomes separated and the state vector of the stationary eq. (27) is written in the form (see (30), (31))

$$(33) \quad \bar{\Phi}(q_k) = \bar{B}(q_k(x)) \exp \left[\frac{i}{\hbar} \bar{S}(q_k) \right],$$

where

$$\bar{S}(q_k) = \int_{\sigma} S(q_k(x)) d\sigma.$$

Since \bar{B} is independent of σ , placing $\bar{B}^2 U(q_k) = \bar{J}(q_k)$, we have the probability conservation in the form (see (20')):

$$(34) \quad \bar{B}^2(q_k) U(q_k) = \bar{B}^2(q_{k,\sigma}) U(q_{k,\sigma}) \frac{d\Sigma_0}{d\Sigma},$$

where $U(q_k)$ is to be understood as velocity in the functional space of the q with components $U_r(q_k)$ ($k=1, 2, \dots, f$) related to $U(q_k)$ by the relation

$$U(q_k) = \left[\sum_r U_r^2(q_k) \right]^{\frac{1}{2}}$$

it should also be noted that in general (see eq. (20) and Part III (30)) the component does not depend only from the correspondent q_r but from all q_k . For free fields we have instead $U_k = U_k(q_k)$, $k=1, 2, \dots, f$.

Following the procedure relative to systems with a finite number of degrees of freedom (⁷), we impose the condition that $\bar{\Phi}(q_k)$ (and thus $\bar{\Psi}(q_k, \sigma)$) may have only one value, then if $\bar{B}(q_k)$ and $\bar{S}(q_k)$ are not single valued for $q_h = q_h^*$, the functional $\bar{\Phi}(q_h^*)$ continues to be single valued if

$$(35) \quad \Delta \bar{S} = \hbar \left[n + \frac{i}{2\pi} \Delta \log \bar{B} \right], \quad n = 0, 1, 2, \dots$$

where Δ indicates the difference between any two values of functionals \bar{S} , $\log \bar{B}$ for $q_h = q_h^*$. Eq. (35) may be easily verified by means of the differentiation of (33) and keeping present that if \bar{B} is always single valued $\Delta \bar{S} = nh$.

We can write (35), equivalent to (36), in the following form

$$(36) \quad \oint \frac{\partial \bar{S}}{\partial q_k} dq_k = \hbar \left[n + \frac{i}{2\pi} \oint \frac{\partial \log \bar{B}}{\partial q_k} dq_k \right], \quad n = 0, 1, 2, \dots$$

where the cyclic integrals are extended along some closed lines in functional space (⁷). Taking into account the probability conservation in the form of (34),

proceeding cautiously, we may extend Keller's arguments for systems with a finite number of degrees of freedom, to systems with an infinite number of degrees of freedom. We will now summarize these considerations, extending the definition to the actual case and citing the essential conclusions.

From (34) we note that if $U d\Sigma$ is infinitesimal in one point in functional space, then \bar{B}^2 is infinite at this point. These are called caustic points.

We are looking for solutions of the type:

$$(37) \quad \Phi(q_k) = \sum_r^M \bar{B}_r(q_k) \exp \left[\frac{i}{\hbar} \bar{S}_r(q_k) \right],$$

to a stationary problem (27) where \bar{B}_r , \bar{S}_r (which may be complex) are considered to be like branches of many valued functionals \bar{B} and \bar{S} given by (33). We introduce for q_k a space C which is M -sheeted such that in each sheet, *e.g.*, r , $\partial \bar{S}_r / \partial q_k$, $\partial \log \bar{B}_r / \partial q_k$ are single valued, then it results that the sheets of C space are joined together in points where

$$\frac{\partial \bar{S}_r}{\partial q_k} = \frac{\partial \bar{S}_s}{\partial q_k},$$

and thus (see (34))

$$(38) \quad \frac{\partial \log \bar{B}_r}{\partial q_k} = \frac{\partial \log \bar{B}_s}{\partial q_k},$$

and the paths of the cyclic integrals in (36), are taken over closed lines in the C -space, such that one satisfies (38), while for other properties these lines may be arbitrary. Under these conditions the expressions of the state vector in (37) and (33) may be considered equivalent.

If we consider a caustic of the type $d\Sigma = 0$ ($U \neq 0$), in this there is an envelope of the family of trajectories, thus $\partial \bar{S} / \partial q_k$ is multiple valued near this surface and the caustic may be considered as the locus of points in which two different branches of $\partial \bar{S} / \partial q_k$ become equal, *i.e.*, the locus where the C -space is joined to itself.

Then too, the caustic $U(q_k) = 0$ ($d\Sigma \neq 0$) is part of the locus where the different branches of the C -space are joined together. In fact, if $U(q_k) = 0$ is so, then $U_r(q_k) = 0$ must follow and taking into account the general form of the component of functional velocity (see (21)), we see that in this case the conjugate momenta on σ , $p_{kv} n_v$, (that coincide with $\partial S / \partial q_k$) are either zero near the caustic (free fields), or they change sign (coupled fields); *i.e.*, this type of caustic is still a locus of points in which two branches of $\partial \bar{S} / \partial q_k$ (and thus $\partial \bar{S} / \partial q_k$) are joined together.

In the caustic the $d\Sigma = 0$, \bar{B} becomes infinite and furthermore, the caustic

must be crossed by a path which goes from one sheet to another of C -space; in general the phase of \bar{B} is retarded by $m(\pi/2)$ over a path that intersects a caustic where the trajectory tube loses m dimensions. Analogously, in the caustic $U(q_k) = 0$ the phase of \bar{B} is still retarded by $m(\pi/2)$ where m is the number of conjugate momenta (integrated over σ) which change sign. In both cases $\log \bar{B}$ varies by $-im\pi/2$ and therefore we have

$$(39) \quad \frac{i}{2\pi} \Delta \log \bar{B} = \frac{i}{2\pi} \oint \frac{\partial \log \bar{B}}{\partial q_k} dq_k = \frac{m}{4}.$$

If we suppose that it is possible to interchange the cyclic integration with the integration on the space-like surface σ , we may write (36) in the form

$$(40) \quad \int_{\sigma} d\sigma \oint \frac{\partial S}{\partial q_k} dq_k = h \left(n + \frac{m}{4} \right).$$

Thus (40) represents the quantum conditions in field theory for separable or unseparable systems; the partial derivatives $\partial S / \partial q_k$ are given by Jacobi's equation. Eq. (40) will be the basis for later applications.

For the sake of completeness we will show ⁽¹⁰⁾ how this relation is connected to the solution in power series of h of the problem contained in the «time-independent» equation of motion (27).

For the sake of simplicity we consider the electromagnetic field in the presence of a given current distribution; the classical hamiltonian function is given by (see Part II (4)):

$$(41) \quad H = -\frac{e^2}{8} P_{\mu\nu} P_{\mu\nu} - \frac{4}{c} J_{\mu} A_{\mu},$$

and the equation of motion in the Schrödinger representation is

$$(42) \quad \left\{ -\frac{e^2}{8} \hbar^2 \frac{\delta^2}{\delta A_{\mu}^2(x)} - \frac{4}{c} J_{\mu} A_{\mu} \right\} \bar{\Psi}(A_{\mu}, J_{\mu}, \sigma) = i\hbar c \frac{\delta \bar{\Psi}}{\delta \sigma(x)};$$

for solving (41), we consider $\bar{\Psi}$ as a functional of the form (see (29), (30))

$$(43) \quad \bar{\Psi}(A_{\mu}, J_{\mu}, \sigma) = \bar{R}(\sigma) \exp \left[\frac{i}{\hbar} \int_{\sigma}^{\mu} \bar{y}_{\mu} (A_{\lambda}, J_{\lambda}) dA_{\mu} \right],$$

⁽¹⁰⁾ E. PERSICO: *Fondamenti della meccanica atomica* (1936).

with

$$(44) \quad \bar{y}_\mu = \int_{\sigma} y_\mu(A_\lambda, J_\lambda) d\sigma,$$

i.e., we use the well known Riccati transformation.

Equation (41) becomes:

$$(45) \quad \left\{ -\frac{ic^2}{8} \hbar \frac{\partial y_\mu}{\partial A_\mu} + \frac{c^2}{8} y_\mu^2 - \frac{4}{c} J_\mu A_\mu \right\} \bar{R}(\sigma) = \frac{i\hbar c \delta \bar{R}(\sigma)}{\delta \sigma(x)},$$

and it can be separated in a « time-dependent » part which gives solution (29), and in a « time-independent » part

$$(46) \quad -\frac{ic^2}{8} \hbar \frac{\partial y_\mu[A_\lambda(x)]}{\partial A_\mu(x)} - \frac{c^2}{8} y_\mu^2[A_\lambda(x)] - \frac{4}{c} J_\mu(x) A_\mu(x) = T(x),$$

where x is the generic point of space-like surface σ where the functional \bar{y}_μ , given by (43), is defined, and $T(x)$ is a point invariant function.

To integrate the equation (45), we suppose that it can be separated for each components of the potentials A_μ . For this we place, on the right hand side of the equation (45), a function $\theta_\mu(x)$, such that once we have established the eigenvalues of $\theta_\mu(x)$, we can reconstruct the invariant function $T(x)$ in such a way that $\int T(x) d\sigma$ may have a physical significance, now the equation may be written as:

$$(47) \quad -\frac{ic^2}{8} \hbar \frac{dy_\mu}{dA_\mu} + \frac{c^2}{8} y_\mu^2 - \frac{4}{c} J_\mu A_\mu = \theta_\mu,$$

and suppose that \bar{y}_μ depends only on A_μ and J_μ . Now we can search for the solution of (47) using the development

$$(48) \quad y_\mu = y_{\mu 0} + \frac{\hbar}{i} y_{\mu 1} + \left(\frac{\hbar}{i} \right)^2 y_{\mu 2} + \dots$$

By putting (48) into (47) and equating the same powers of \hbar , it may be easily seen that we obtain the recurrence equation:

$$(49) \quad \begin{cases} y_{\mu 0}^2 = \frac{8}{c^2} \left(\theta_\mu(x) + \frac{4}{c} J_\mu(x) A_\mu(x) \right), \\ y_{\mu 1} = -\frac{1}{2y_{\mu 0}} \frac{dy_{\mu 0}}{dA_\mu}, \\ y_{\mu 2} = -\frac{1}{2y_{\mu 0}} \left(y_{\mu 1}^2 + \frac{dy_{\mu 1}}{dA_\mu} \right), \\ \dots \end{cases}$$

Now if we set

$$(50) \quad p_\mu(A_\mu(x), J_\mu(x)) = \sqrt{\frac{8}{c^2} \left(\theta_\mu(x) + \frac{4}{c} J_\mu(x) A_\mu(x) \right)},$$

we have

$$y_{\mu 0} = \pm p_\mu$$

and we can write for the first two terms of (47):

$$(51) \quad \begin{cases} y_{\mu a} = p_\mu(A_\mu, J_\mu) + \frac{i\hbar}{2p_\mu[A_\mu(x)]} \frac{dp_\mu(A_\mu, J_\mu)}{dA_\mu(x)}, \\ y_{\mu b} = -p_\mu(A_\mu, J_\mu) + \frac{i\hbar}{2p_\mu[A_\mu(x)]} \frac{dp_\mu(A_\mu, J_\mu)}{dA_\mu(x)}. \end{cases}$$

With this approximation (for each μ), the general integral of equation (41) can be written:

$$(52) \quad \bar{\Psi}(A_\mu, J_\mu, \sigma) = \bar{R}(\sigma) \bar{B}(p_\mu) \cdot \left\{ c_{\mu a} \exp \left[\frac{i}{\hbar} \int_\sigma^{\bar{A}_\mu^*} d\sigma \int_{\bar{A}_\mu^*}^{\bar{A}_\mu^*} p_\mu(A_\mu, J_\mu) dA_\mu \right] + c_{\mu b} \exp \left[-\frac{i}{\hbar} \int_\sigma^{\bar{A}_\mu^*} d\sigma \int_{\bar{A}_\mu^*}^{\bar{A}_\mu^*} p_\mu(A_\mu, J_\mu) dA_\mu \right] \right\},$$

where $C_{\mu a}$ and $C_{\mu b}$ are constants, \bar{A}_μ^* is some arbitrary point of functional space, and the amplitude of the state vector (apart from $\bar{R}(\sigma)$) is given by the real functional:

$$(53) \quad \bar{B}(p_\mu) = \exp \left[-\int d\sigma \log p_\mu[A_\mu(x), J_\mu(x)] \right].$$

Solution (52) is of the form of (37) and if we impose the condition that the functional $\bar{\Psi}$ must be single valued, we obtain for each μ the quantum condition (see (40))

$$(54) \quad \int_\sigma d\sigma \oint p_\mu[A_\mu(x), J_\mu(x)] dA_\mu = \hbar \left(n + \frac{1}{2} \right),$$

where the limits of the cyclic integral are given by the condition that p_μ must be real. Such a condition in fact, ensures that two functions p_μ change sign, so that (40) we have $m = 2$. Furthermore the function p_μ is equivalent to the function $\partial S / \partial q_k$ of the general formula (40). In fact, with the hamiltonian (41), and with the function V_r of the type (31), Jacobi's equation (23)

becomes

$$(55) \quad \frac{c^2}{8} \left(\frac{\partial S}{\partial A_\mu} \right)^2 - \frac{4}{c} J_\mu A_\mu - \theta_\mu(x) = 0,$$

from which it follows immediately that for each μ we have $p_\mu = \partial S / \partial A_\mu$. Finally we must remember that the « separation » of $T(x)$ is arbitrary, so that from the relation (54) we must construct a relation independent of μ in which $T(x)$ is an invariant. This will be done in some of the following applications.

Now we may conclude that, as in the case of systems of a finite number of degrees of freedom, it is possible to establish quantum conditions for stationary states. They pertain to some integral condition of the function $T(x)$ and, apart from other eventual inner degrees of freedom, they are the correct integral eigenvalues of the problem contained in eq. (27), if we desire to know $\int T(x) d\sigma$; in fact they are obtained by means of topological conditions. The relations (51), (52), (54), (55) prove that when we solve equation (27) in series of powers of \hbar , (up to the first order on \hbar) imposing the conditions of uniqueness for the amplitude and the phase, we obtain the correct quantum conditions, i.e., we solve the problem of the integral eigenvalues, but the state vector remains approximate.

We recall finally that from (27) it should be possible to find the function $T(x)$.

4. - On the physical meaning of quantum conditions.

In order to obtain the physical meaning of quantum conditions it will be useful to consider some simple applications of formula (40). First we will consider the simplest cases of free boson and fermion fields.

a) *Scalar meson field.* - From the lagrangian

$$(56) \quad L = -\frac{1}{2} \left(\frac{\partial \varphi}{\partial x_\nu} \right)^2 - \frac{1}{2} \mu^2 \varphi^2, \quad \mu = \frac{m_0 c}{\hbar},$$

with the position for conjugate momenta $P_\nu = -4c(\partial \varphi / \partial x_\nu)$, we have the hamiltonian

$$(57) \quad H = -\frac{c^2}{8} P_\nu^2 + 2\mu^2 \varphi^2,$$

and if we consider Jacobi's function of the type (see (31))

$$(58) \quad V_\nu = n_\nu \left(S(\varphi) - \int_{\tau_0}^{\tau} T(x) d\tau \right),$$

Jacobi's equation (23) relative to (57) becomes

$$(59) \quad \frac{e^2}{8} \left(\frac{\partial S}{\partial \varphi} \right)^2 + 2\mu^2 \varphi^2 = T(x).$$

This equation for each point x of space-like surface is the equation of a harmonic oscillator in functional space. It is equivalent to the equation

$$(60) \quad \frac{\partial S}{\partial \varphi} = \pm \frac{2}{e} \sqrt{2T - 4\mu^2 \varphi^2},$$

if we impose the condition of reality on (60), *i.e.*, we suppose that

$$(61) \quad -\frac{\sqrt{2T}}{2\mu} \leq \varphi \leq \frac{\sqrt{2T}}{2\mu},$$

we have $\oint (\partial S / \partial \varphi) d\varphi = 2\pi T(x) / \mu e$, and because two momenta change their signs $m=2$ in (40), we now have the formula:

$$(62) \quad \int_{\sigma} T_n(x) d\sigma = (n + \frac{1}{2}) m_0 c^2, \quad n = 0, 1, 2, \dots$$

where we put the index n on the left hand side.

This is clearly the mass spectrum of a free boson field, it agrees with the energy spectrum given by standard field theory ⁽¹¹⁾.

b) Electromagnetic field. - If the lagrangian is $L = -\frac{1}{2} (\partial A_\mu / \partial x_\nu) (\partial A_\mu / \partial x_\nu)$, and placing $P_{\mu\nu} = -4e (\partial A_\mu / \partial x_\nu)$ the hamiltonian for the free electromagnetic field is given by

$$(63) \quad H = -\frac{e^2}{8} P_{\mu\nu} P_{\mu\nu},$$

if we choose Jacobi's function of the type

$$(64) \quad V_v = n_v \left(S(A_\mu) - \int_{\tau_0}^{\tau} T(x) d\tau \right),$$

Jacobi's equation becomes

$$(65) \quad \frac{e^2}{8} \left(\frac{\partial S}{\partial A_\mu} \right)^2 = T(x);$$

for each x it represents a free point in functional space.

⁽¹¹⁾ G. WENTZEL: *Quantum Theory of Fields* (1949).

In this case we may find the quantum conditions in two equivalent ways. We separate equation (65) introducing for each μ a function $\theta_\mu(x)$ and a mass m_0 independent of μ , so that we have the equations

$$(66) \quad \frac{e^2}{8} \left(\frac{\partial S_\mu}{\partial A_\mu} \right)^2 + \frac{2m_0^2 e^2}{\hbar^2} A_\mu^2 = \theta_\mu, \quad \mu = 1, 2, 3, 4,$$

and as for the meson case we obtain the quantum conditions

$$(67) \quad \int_\sigma \theta_{\mu n}(x) d\sigma = (n + \tfrac{1}{2})_\mu m_0 e^2.$$

The functions $\theta_\mu(x)$ must be such that

$$(68) \quad \sum_{\mu=1}^4 \theta_\mu(x) = T(x)$$

and adding up the equations (67) for $\mu = 1, 2, 3, 4$ we must obtain an invariant relation. This means that also the right hand side of (67) must be independent of μ and we obtain:

$$(69) \quad \int_\sigma T(x) d\sigma = 4(n + \tfrac{1}{2}) m_0 e^2.$$

When $m_0 \rightarrow 0$ the vector meson field of equation (66) becomes the electromagnetic field, then we find from (69) the quantum condition

$$(70) \quad \int_\sigma T(x) d\sigma = 0.$$

We can also find this relation starting from (66), with $m_0 = 0$. Following Keller's topological argument we see that the paths of the integration in functional space can be deformed in a point so that the right-hand side of (40) is zero for each μ ; i.e., (70).

c) *Meson field interacting with electromagnetic field.* - The hamiltonian is given, starting from the usual lagrangian, (see Part III (8)), by

$$(71) \quad H = -\frac{e^2}{8} P_{\mu\nu} P_{\mu\nu} - \frac{e^2}{4} P_\nu^* P_\nu + \frac{ie}{\hbar} A_\nu (P_\nu \varphi - P_\nu^* \varphi^*) + 4\mu^2 \varphi^* \varphi - \\ - \frac{e^2}{8} P_{3\nu}^2 + 2\mu^2 \varphi_3^2,$$

where the index 3 refers to the neutral meson field. Changing the complex variables φ^* , φ with the positions

$$(72) \quad \sqrt{2}\varphi = \varphi_1 + i\varphi_2, \quad \sqrt{2}\varphi^* = \varphi_1 - i\varphi_2,$$

where the φ_1 and φ_2 refer to positive and negative charged mesons and calling $P_{1\nu}$, $P_{2\nu}$ the new conjugate momenta, the hamiltonian becomes

$$(73) \quad H = -\frac{e^2}{8} P_{\mu\nu} P_{\mu\nu} - \frac{e^2}{8} (P_{1\nu}^2 + P_{2\nu}^2 + P_{3\nu}^2) - \\ - \frac{e}{\hbar} A_\nu (P_{1\nu} \varphi_2 - P_{2\nu} \varphi_1) + 2\mu^2 (\varphi_1^2 + \varphi_2^2 + \varphi_3^2),$$

from which Jacobi's equation for stationary states is given by

$$(74) \quad \frac{e^2}{8} \left(\frac{\partial S}{\partial A_\mu} \right)^2 + \frac{e^2}{8} \left[\left(\frac{\partial S}{\partial \varphi_1} \right)^2 + \left(\frac{\partial S}{\partial \varphi_2} \right)^2 + \left(\frac{\partial S}{\partial \varphi_3} \right)^2 \right] + \frac{e}{\hbar} \left(\frac{\partial S}{\partial \varphi_2} \varphi_1 - \frac{\partial S}{\partial \varphi_1} \varphi_2 \right) n_\nu A_\nu + \\ + 2\mu^2 (\varphi_1^2 + \varphi_2^2 + \varphi_3^2) = T(x).$$

We neglect for the moment the electromagnetic field, i.e., we consider equation (74) without the first and the third terms. We have therefore the equation of motion of a three dimensional harmonic oscillator.

If we introduce the customary isotopic spin space and we use polar coordinates in this space, the Jacobi equation becomes

$$(75) \quad \frac{e^2}{8} \left\{ \left(\frac{\partial S}{\partial \varrho} \right)^2 + \frac{1}{\varrho^2} \left(\frac{\partial S}{\partial \theta} \right)^2 + \frac{1}{\varrho^2 \sin^2 \theta} \left(\frac{\partial S}{\partial \alpha} \right)^2 \right\} + 2\mu^2 \varrho^2 = T(x).$$

Following the old methods of Sommerfeld's theory⁽¹²⁾ we can separate this equation and we have the three quantum condition

$$(76) \quad \int_\sigma d\sigma \oint \frac{\partial S}{\partial \varrho} d\varrho = n_\varrho h; \quad \int_\sigma d\sigma \oint \frac{\partial S}{\partial \theta} d\theta = s h; \quad \int_\sigma d\sigma \oint \frac{\partial S}{\partial \alpha} d\alpha = m^* h,$$

(all variables ϱ , θ , α have the same «period» and in (40) $m = 0$).

The third equation is obviously a condition for the third component of the isotopic spin (third component of the angular momentum in functional space) and since⁽¹³⁾

$$(77) \quad \frac{e}{\hbar} \int d\sigma \frac{\partial S}{\partial \alpha} = Q,$$

⁽¹²⁾ A. SOMMERFELD: *Atomic Structure and Spectral Lines*, English translation (1923), p. 551 on.

⁽¹³⁾ J. HAMILTON: *The Theory of Elementary Particles* (1959).

is the total charge, and $\partial S/\partial \alpha$ is a constant in isotopic spin space, the third condition is simply

$$(78) \quad Q = m^* e \quad m^* = \dots - 2, -1, 0, 1, 2, \dots$$

Summing up the second and third conditions and introducing a new quantum number $k = s + |m^*|$, we have, from the condition that the total angular momentum K (total isotopic spin) is a constant, the condition

$$(79) \quad \int d\sigma K(x) = k\hbar,$$

where k is an integer positive number or zero.

If we call β the angle between \mathbf{K} and the polar axis we have

$$(80) \quad K_3 = K \cos \beta$$

and (for $K \neq 0$)

$$(81) \quad \cos \beta = \frac{m^*}{k} = 0, \quad \pm \frac{1}{k}, \quad \pm \frac{2}{k}, \quad \dots, \quad \pm 1.$$

Since we have, for the total angular momentum in functional space

$$(82) \quad \left(\frac{\partial S}{\partial \theta} \right)^2 + \frac{1}{\sin^2 \theta} \left(\frac{\partial S}{\partial \alpha} \right)^2 = K^2,$$

Jacobi's equation becomes

$$(83) \quad \frac{e^2}{8} \left\{ \left(\frac{\partial S}{\partial \varrho} \right)^2 + \frac{K^2}{\varrho^2} \right\} + 2\mu^2 \varrho^2 = T,$$

so that the first quantum condition becomes

$$(84) \quad \int_{\sigma} d\sigma \oint \frac{2\sqrt{2}}{e} \left(T - 2\mu^2 \varrho^2 - \frac{e^2}{8} \frac{K^2}{\varrho^2} \right) d\varrho = n_e \hbar.$$

The cyclic integral was worked out by SOMMERFELD⁽¹²⁾, and using (79) we have

$$(85) \quad \int_{\sigma} d\sigma \frac{2\pi T(x)}{\mu c} = (n_e + k) \hbar.$$

Introducing a new quantum number $n = n_e + k$ (which can be zero or an integer and following the usual terminology, we call it the total quantum number), (85) gives

$$(86) \quad \int_{\sigma} T_n(x) d\sigma = nm_0c^2.$$

It is easy to show that the total quantum number n is that of the general formula (40). In fact, following the topological considerations, we can write the quantum conditions in the form

$$(87) \quad \int_{\sigma} d\sigma \oint \left(\frac{\partial S}{\partial \varrho} d\varrho + \frac{S\partial}{\partial \theta} d\theta + \frac{\partial S}{\partial \alpha} d\alpha \right) = nh,$$

and recalling that the total angular momentum in functional space is constant, i.e., (79), we obtain:

$$(88) \quad \int_{\sigma} d\sigma \oint \frac{\partial S}{\partial \varrho} d\varrho = (n - k)h = n_e h,$$

and because we want solutions in which $\int_{\sigma} T(x) d\sigma$ is a non-negative number, it follows that

$$(89) \quad k \leq n.$$

In other words, the methods of quantization of a doubly degenerate system gives us, for a value of the charge, a mass spectrum; the conditions (81) (89) ensure that if the value of the mass is nm_0c^2 , the value of the charge is m^*e with $m^* \leq n$. It relates mass, charge (third component of isotopic spin) and isotopic spin. In this case we do not have the term $\frac{1}{2}m_0c^2$ of (62).

If we also consider in (74) the contribution of the electromagnetic field, it should be possible to avoid one of these degenerations. The method is the same as that used in previous cases, but in this one, we obtain some integral relation, from which it is impossible to get directly the value of $\int T(x) d\sigma$. By using the mean value theorem we can obtain certain terms which are corrections for the mass of charged mesons, they behave as R^{-1} where R is the radius of the sphere V to which we extend the previous integral, and in order to get the mass difference $c^2 \Delta m_{\pm} \cong 10 m_e c^2$; it is necessary to suppose that R should be of the order of five times the Compton wave length of the π -meson. As we said in the introduction, this hypothesis may be controlled using the point-solution of equation (27).

d) *Spinor field*. — From the hamiltonian (15), if we neglect the first three terms, we obtain for Jacobi's equation of the spinor field

$$(90) \quad \frac{k_0}{4\hbar c} \frac{\partial S}{\partial \bar{\varphi}} \frac{\partial S}{\partial \varphi} + 4m_0 c^2 \bar{\varphi} \varphi + \frac{2e^2}{k_0^2} (\bar{\varphi} \gamma_\mu \gamma_\nu \varphi)^2 = T(x).$$

Since we use only the first two components of the spinors, it results that

$$(91) \quad \bar{\varphi}_i = \varphi_i^* \quad l = 1, 2$$

and if we neglect for the moment the contribution of the spin term in (90), with a position analogous to (72), and then using plane polar co-ordinates in functional space, we obtain for each spin direction

$$(92) \quad \frac{k_0}{8\hbar c} \left[\left(\frac{\partial S}{\partial \varrho} \right)^2 + \frac{1}{\varrho^2} \left(\frac{\partial S}{\partial \alpha} \right)^2 \right] + 2m_0 c^2 \varrho^2 = T(x),$$

i.e., Jacobi's equation for an harmonic oscillator in the plane. As for the meson case we have the quantum condition for the third component of «angular momentum»

$$(93) \quad 2\pi \int_{\sigma} d\sigma \frac{\partial S}{\partial \alpha} = m^* \hbar,$$

where $m^* = n_+ - n_-$ is the difference between the numbers of positive and negative charges and the total charge is $Q = m^* e$.

We also have a quantum condition analogous to (86), but according to Pauli's exclusion principle, the spin direction being fixed, we must write:

$$(94) \quad \int_{\sigma} T(x) d\sigma = (n_+ + n_-) m_0 a^2, \quad n_+ = 0.1, \quad n_- = 0.1,$$

so that in (93) m^* can have only the values $-1, 0, 1$.

We consider now the contribution of the spin term in (90). The spinors having two components, we can choose a representation for the spin operator such that it results:

$$(95) \quad \sigma_{12} \varphi = \begin{vmatrix} \varphi_1 \\ -\varphi_2 \end{vmatrix}, \quad \sigma_{23} \varphi = \begin{vmatrix} \varphi_2 \\ \varphi_1 \end{vmatrix}, \quad \sigma_{31} \varphi = i \begin{vmatrix} \varphi_2 \\ -\varphi_1 \end{vmatrix},$$

and we obtain

$$(96) \quad (\bar{\varphi} \gamma_\mu \gamma_\nu \varphi)^2 = -\{(\bar{\varphi} \sigma_{12} \varphi)^2 + (\bar{\varphi} \sigma_{23} \varphi)^2 + (\bar{\varphi} \sigma_{13} \varphi)^2\} = -(\varphi_1^* \varphi_1 - \varphi_2^* \varphi_2)^2.$$

If we assume states of definite spin we have one of two possibilities

$$(97) \quad (\bar{\varphi} \gamma_{\mu} \gamma_3 \varphi)^2 = \begin{cases} -(\varphi_1^* \varphi_1)^2, \\ -(\varphi_2^* \varphi_2)^2, \end{cases}$$

and, for each direction of the spin, from (90) we obtain

$$(98) \quad \frac{k_0}{8\hbar c} \left[\left(\frac{\partial S}{\partial \varrho} \right)^2 + \frac{1}{\varrho^2} \left(\frac{\partial S}{\partial \alpha} \right)^2 \right] + 2m_0 c^2 \varrho^2 - \frac{e^2}{2k_0^2} \varrho^4 = T(x),$$

which is (92) with the spin term. As in the previous case we assume that $\partial S / \partial \alpha$ is constant in functional space and that the term of the spin in ϱ^4 is a small correction in (98). Therefore we have for the radial quantum condition

$$(99) \quad \int_{\sigma} d\sigma \oint \frac{\partial S}{\partial \varrho} d\varrho = n_e h = -2\pi \int_{\sigma} \left[\frac{\partial S}{\partial \alpha} + \frac{e^2 \hbar}{64 m_0^3 c^4} \left(\frac{\partial S}{\partial \alpha} \right)^2 \right] d\sigma + \\ + 2\pi \int_{\sigma} \frac{\hbar}{m_0 c^2} \left(T + \frac{3e^2 \hbar^2}{64 m_0^4 c^6} T^2 \right) d\sigma,$$

where the cyclic integral was worked out by SOMMERFELD (see loc. cit.).

From this relation, if we suppose $e^2 = 0$ (i.e., we avoid the spin contribution), we obtain immediately (94) and (93). If we have one electron, since $T(x)$ is positive and regular (see (61) and see (94))

$$\frac{1}{m_0 c^2} \int_{\sigma} T(x) d\sigma = 1,$$

it follows that for the unit volume:

$$\frac{1}{m_0 c^2} T(x) < 1,$$

from which

$$T^2(x) < m_0 c^2 T(x)$$

and we can write for the last integral in (99):

$$(100) \quad \frac{2\pi \hbar}{m_0 c^2} \int_{\sigma} \left(T + \frac{3e^2 \hbar^2}{64 m_0^4 c^6} T^2 \right) d\sigma < \frac{2\pi \hbar}{m_0 c^2} \int_{\sigma} \left(1 + \frac{3e^2 \hbar^2}{64 m_0^3 c^4} \right) T d\sigma.$$

Putting the numerical values in the coefficient of T we find that the correction to $\int T d\sigma$, (*i.e.*, on the mass), is less than 10^{-33} . By means of the same procedure it is possible to find that the correction to $\int (\partial S / \partial \alpha) d\sigma$ (*i.e.*, to the charge) is less than 10^{-33} . Having considered the above, we can still consider the relation (99) as that leading to (93) and (94) but with a small correction to mass and charge.

From all of the above considerations it follows that, at least for free fields, $\int T(x) d\sigma$ have the meaning of mass (rest energy in our unities). Functional formalism gives us some indications on the structure of functional space in field theory. These statements will be the basis for further considerations.

* * *

The author wishes to thank the Consiglio Nazionale delle Ricerche for financial support; the Conference Board of Associated Research Councils, Committee on International Exchange of Persons, for the travel grant; and Professor ROBERT SERBER for the kind interest shown in this work.

RIASSUNTO

Si applica il formalismo funzionale covariante all'equazione spinoriale di Feynman-Gell-Mann. Si estende l'approssimazione al primo ordine del metodo W.K.B. alla teoria dei campi dando le corrispondenti condizioni quantiche. Applicando questi risultati ai campi liberi si trovano delle relazioni fra i vari numeri quantici nella teoria dei campi. Per campi accoppiati si hanno indicazioni per una ulteriore trattazione del problema.

Theoretical Analysis of Absorption of Slow π^- -Mesons in Light Nuclei.

P. AMMIRAJU and S. N. BISWAS

Tata Institute of Fundamental Research - Bombay

(Ricevuto il 6 Giugno 1960)

Summary. — An analysis of the π^- -absorption in light nuclei is presented assuming that the absorption reaction proceeds via the two nucleon π^- -capture mode *viz.*, through the reaction $\pi^- + (N+N^0) \rightarrow N^0+N^0$. Assuming the charge independence of pion-nucleon interaction and unitarity and time-reversal invariance of the S -matrix, the two nucleon π^- -capture reaction cross-sections have been written in terms of the nucleon-nucleon phase shifts. The parameters that enter into the theory are determined from the experimental observation of OZAKI *et al.* Using the principle of detailed balance, a check has been made of the parameters with meson production data at threshold. It is further shown that the two nucleon model provides a consistent explanation of the absorption phenomena of the π^- -meson in helium and carbon.

1. — Introduction.

The absorption of very slow π^- -mesons by complex nuclei has considerable theoretical interest for the problem of nuclear structure. The sudden release of the rest energy of the pion (~ 140 MeV) inside the nucleus and the subsequent distribution of the excitation energy among the nucleons affords a powerful tool to probe the interior of the nucleus. For a meson to be absorbed in the nucleus, one needs at least the participation of two nucleons (*i.e.* a hard scattering of two nucleons) to share the rest mass-energy of the pion in order to satisfy the conservation laws. This requirement has an important bearing on the nuclear correlations inside the nucleus ⁽¹⁾.

⁽¹⁾ K. A. BREUCKNER, R. J. EDEN and R. C. FRANCIS: *Phys. Rev.*, **98**, 1455 (1955).

For a proper understanding of the nuclear structure, one has to establish some relationship between the extreme viewpoints as exemplified in the shell model and the liquid drop model. The simple shell model completely ignores the strong short-range nucleon-nucleon interactions while in the liquid drop model one assumes such strong interactions among the nucleons that any excitation energy given to the nucleus is more or less equally shared among the nucleons. There is ample evidence to show that neither of these viewpoints is correct. In particular the absorption of pions at rest by complex nuclei provides one such type of evidence to show that indeed the nucleons in the nucleus are highly correlated. We will show, further on, that π^- -meson absorption in a complex nucleus essentially takes place by the participation of two nucleons (either a proton-proton pair or a neutron-proton pair) ⁽²⁾. This is in conformity with the usual picture of the short range nucleon-nucleon forces.

Early work ⁽³⁾ on the π^- -meson absorption by nuclei was done using nuclear emulsions which suffer from the disadvantage that the events cannot be unambiguously separated because of the composite mixture of light and heavy elements in the emulsion. Subsequently ⁽⁴⁾, a series of measurements on the absorption of very slow π^- -mesons in light elements has been done using a diffusion cloud chamber filled consecutively with helium, ethylene (carbon) and nitrogen. A detailed analysis of the complex reactions involved have been analysed and interpreted on the basis of nucleon pair capture of the pion. Recently, OZAKI *et al.* ⁽⁵⁾ reported measurements on the absorption of π^- -mesons on carbon and aluminium using counter technique and come to the same conclusions as discussed in reference ⁽⁴⁾. We shall give below a theoretical analysis of the mechanism of the absorption of π^- -meson by light nuclei using the pair absorption model and charge-independence of the pion-nucleon interaction.

2. — Theory of the Isospin analysis.

A negative pion of several MeV energy brought to rest in matter is quickly bound in a low lying mesonic orbit by the Coulomb field of the nucleus. In these states ($1s$, $2p$), the meson is very close to the nucleus and the strong

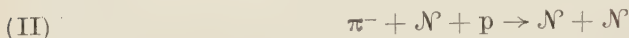
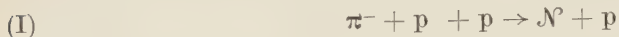
⁽²⁾ This model was first suggested by K. A. BREUCKNER, R. SERBER and K. M. WATSON: *Phys. Rev.*, **81**, 575 (1951); **84**, 258 (1951); D. H. PERKINS: *Phil. Mag.*, **40** 601 (1949). Later also discussed by G. PUPPI, V. DE SABBATA and V. MANARESI: *Nuovo Cimento*, **10**, 1704 (1953).

⁽³⁾ M. G. K. MENON, H. MUIRHEAD and O. ROCHAT: *Phil. Mag.*, **41**, 583 (1950); C. J. BROWN and I. S. HUGHES: *Phil. Mag.*, **2**, 777 (1957).

⁽⁴⁾ P. AMMIRAJU and L. M. LEDERMAN: *Nuovo Cimento*, **4**, 283 (1956).

⁽⁵⁾ S. OZAKI, R. WEINSTEIN, G. GLASS, E. LOH, L. NIEMALA and A. WATTENBERG: *Phys. Rev. Lett.*, **4**, 533 (1960).

Yukawa interaction ($\pi^- + p \rightarrow \mathcal{N}$) will quickly annihilate it. To conserve momentum and energy, we assume that the capture takes place essentially by the following basic reactions



capture by an $\mathcal{N}\mathcal{N}$ pair is precluded by charge conservation. We will, further, assume in the following the charge independence hypothesis (*i.e.*, the interaction is independent of the Z -component of T , the total isotopic spin).

The system ($\pi^- + p + p$) exists in the state of total isotopic spin $T = 2, 1$, and 0 while the two nucleons are in the state of total isotopic spin $T' = 1$, and also in the state $T = 1$ when the two nucleon system has $T' = 0$. We will denote by $\chi_2^{(1)}$, $\chi_1^{(1)}$, and $\chi_0^{(1)}$ the states corresponding to $T = 2, 1$ and 0 respectively when the two nucleon system is in $T' = 1$ and by $\chi_1^{(0)}$ for $T = 1$ with the two nucleon system being in $T' = 0$. We, then, have the following results,

$$(1) \quad \chi_2^{(1)} = \frac{1}{\sqrt{6}} \left[\pi^- pp + 2 \left(\frac{p\mathcal{N} + \mathcal{N}p}{\sqrt{2}} \right) \pi^0 + \mathcal{N}\mathcal{N}\pi^+ \right],$$

$$(2) \quad \chi_1^{(1)} = \frac{1}{\sqrt{2}} [\pi^- pp - \mathcal{N}\mathcal{N}\pi^+],$$

$$(3) \quad \chi_0^{(1)} = \frac{1}{\sqrt{3}} \left[\pi^- pp - \left(\frac{p\mathcal{N} + \mathcal{N}p}{\sqrt{2}} \right) \pi^0 + \mathcal{N}\mathcal{N}\pi^+ \right].$$

For all the above states $T_z = 0$ because on the right hand side of reaction (I) only $T_z = 0$ is allowed. From (1), (2) and (3), we can write the physically interesting state ($\pi^- pp$) in terms of the eigenstates of total T .

Thus,

$$(4) \quad \pi^- pp = \frac{1}{\sqrt{6}} \chi_2^{(1)} + \frac{1}{\sqrt{2}} \chi_1^{(1)} + \frac{1}{\sqrt{3}} \chi_0^{(1)}.$$

It is clear that the total isotopic spin state of the two nucleons is denoted by the superscript while the subscript is the one that is conserved in the over all interaction.

Similarly, the ($\pi^- \mathcal{N}p$) state can be expressed in terms of the eigenstates of the total isotopic spin as follows:

$$(5) \quad \chi_1^{(1)} = \frac{1}{\sqrt{2}} \left[\pi^- \left(\frac{p\mathcal{N} + \mathcal{N}p}{\sqrt{2}} \right) + \mathcal{N}\mathcal{N}\pi^0 \right],$$

$$(6) \quad \chi_1^{(1)} = \frac{1}{\sqrt{2}} \left[\pi^- \left(\frac{p\mathcal{N} - \mathcal{N}p}{\sqrt{2}} \right) - \mathcal{N}\mathcal{N}\pi^0 \right],$$

$$(7) \quad \chi_1^{(0)} = \pi^- \left(\frac{p\mathcal{N} - \mathcal{N}p}{\sqrt{2}} \right).$$

By simple algebra, we get the following expressions for the states $(\pi^- \mathcal{N} p)$ and $(\pi^- p \mathcal{N})$

$$(8) \quad \pi^- p \mathcal{N} = \frac{1}{2} (\chi_2^{(1)} + \chi_1^{(1)}) + \frac{1}{\sqrt{2}} \chi_1^{(0)},$$

$$(9) \quad \pi^- \mathcal{N} p = \frac{1}{2} (\chi_2^{(1)} + \chi_1^{(1)}) - \frac{1}{\sqrt{2}} \chi_1^{(0)}.$$

Let us denote the final state of reaction (I) by $1/\sqrt{2}(\varphi_1 + \varphi_0)$ and of reaction (II) by φ , where

$$\varphi_1 = \frac{p \mathcal{N} + \mathcal{N} p}{\sqrt{2}} \quad \text{and} \quad \varphi_0 = \frac{p \mathcal{N} - \mathcal{N} p}{\sqrt{2}}.$$

The S -matrix element of reaction (I) is given by

$$(10) \quad \langle \pi^- p p | S | \mathcal{N} p \rangle = \left\langle \frac{1}{\sqrt{6}} \chi_2^{(1)} + \frac{1}{\sqrt{2}} \chi_1^{(1)} + \frac{1}{\sqrt{3}} \chi_0^{(1)} | S | \frac{1}{\sqrt{2}} (\varphi_1 + \varphi_0) \right\rangle.$$

Hence from (10) we have

$$(11) \quad \langle \pi^- p p | S | \mathcal{N} p \rangle = \frac{1}{\sqrt{6}} \langle \chi_0^{(1)} | S | \varphi_0 \rangle + \frac{1}{2} \langle \chi_1^{(1)} | S | \varphi_1 \rangle.$$

From time reversal invariance of S -matrix ⁽⁶⁾, we can easily write the above matrix-elements as

$$(12) \quad \langle \pi^- p p | S | \mathcal{N} p \rangle = \frac{1}{\sqrt{6}} |b_0^{(1)}| \exp[i\delta_0] + \frac{1}{2} |b_1^{(1)}| \exp[i\delta_1],$$

where $\langle \varphi_0 | S_0 | \varphi_0 \rangle \approx \exp[i\delta_0]$ and $\langle \varphi_1 | S_0 | \varphi_1 \rangle \approx \exp[i\delta_1]$ with δ_0 and δ_1 being the singlet and triplet nucleon-nucleon phase shifts and $|b_0^{(1)}|$ and $|b_1^{(1)}|$ being the corresponding amplitudes and S_0 denotes the diagonal part of the S -matrix.

The cross-section $\sigma(\pi^- + p + p \rightarrow \mathcal{N} p)$ denoted by $\sigma(\text{pp})$ is given by

$$(13) \quad \sigma(\text{pp}) \approx \frac{1}{6} |b_0^{(1)}|^2 + \frac{1}{4} |b_1^{(1)}|^2 + \frac{1}{\sqrt{6}} |b_0^{(1)}| |b_1^{(1)}| \cos(\delta_0 - \delta_1),$$

similarly we can evaluate the cross-section $\sigma(\pi^- + \mathcal{N} + p \rightarrow \mathcal{N} \mathcal{N}) = \sigma(\mathcal{N} p)$ as

⁽⁶⁾ K. M. WATSON: *Phys. Rev.*, **95**, 228 (1954).

follows:

$$(14) \quad \langle \pi \mathcal{N} p | S | \mathcal{N} \mathcal{N} \rangle = \langle \frac{1}{2}(\chi_2^{(1)} + \chi_1^{(1)}) - \frac{1}{2}\chi_1^{(0)} | S | \varphi_1 \rangle = \\ = \frac{1}{2}\langle \chi_1^{(1)} | S | \varphi_1 \rangle - \frac{1}{2}\langle \chi_1^{(0)} | S | \varphi_1 \rangle,$$

$$(15) \quad \langle \pi p \mathcal{N} | S | \mathcal{N} \mathcal{N} \rangle = \frac{1}{2}\langle \chi_1^{(1)} | S | \varphi_1 \rangle + \frac{1}{2}\langle \chi_1^{(0)} | S | \varphi_1 \rangle.$$

Using Watson's theorem ⁽⁶⁾ we can write similarly

$$\langle \chi_1^{(1)} | S | \varphi_1 \rangle = [b_1^{(1)}] \exp [i\delta_1],$$

$$\langle \chi_1^{(0)} | S | \varphi_1 \rangle = [b_1^{(0)}] \exp [i\delta_1].$$

Then,

$$(16) \quad \sigma(\mathcal{N}p) \approx \frac{1}{8} [|b_1^{(1)}|^2 + |b_1^{(0)}|^2].$$

For normalization, we take one of the parameters $|b_1^{(1)}|$ to unity, *i.e.* we will determine $|b_1^{(0)}|$ and $|b_0^{(1)}|$ in units of $|b_1^{(1)}|$. We then have

$$(17) \quad \sigma(\mathcal{N}p) \approx \frac{1}{8} [1 + |b_1^{(0)}|^2],$$

$$(18) \quad \sigma(pp) \approx \frac{1}{6} |b_0^{(1)}|^2 + \frac{1}{4} + \frac{1}{6} |b_0^{(1)}| \cos (\delta_0 - \delta_1).$$

We now take the ratio

$$(18a) \quad \sigma(\mathcal{N}p)/\sigma(pp) = 5$$

consistent with the experimental results of OZAKI *et al.* ⁽⁵⁾ on the observation of the $\mathcal{N}\mathcal{N}$ and $\mathcal{N}p$ coincidences from the π^- -absorption in carbon. Assuming the conservation of angular momentum and parity in (I) and (II), the nucleons are in the triplet P -state, we have $\delta_0 - \delta_1 \approx 18^\circ$ ⁽⁷⁾ and $\cos (\delta_0 - \delta_1) \approx .95$. From (17), (18) and (18a) we get then the following relationship among the $|b|$'s

$$(19) \quad |b_1^{(1)}|^2 = \frac{40}{6} |b_0^{(1)}|^2 + 9 + \frac{40}{\sqrt{6}} |b_0^{(1)}|$$

one easily sees that a possible solution consistent with the experiment is given by

$$|b_1^{(1)}| = 1, \quad |b_1^{(0)}| = 3 \quad \text{and} \quad |b_0^{(1)}| = 0.04$$

we will now show that these values of the parameters are consistent with the meson production cross-sections at threshold.

⁽⁷⁾ *High Energy Nuclear Physics Conference, Rochester (1957)*, p. III-12.

3. - Relationship between the absorption and production cross-sections of the pion with the nucleons⁽⁴⁾.

Consider the reaction (I)

$$\pi^- + p + p \rightarrow N + p.$$

This corresponds to the inverse production reaction

$$N + p \rightarrow p + p + \pi^-.$$

Assuming this latter reaction for 1S_0 state of the final protons and S -state of the emitted meson, the initial state of the two nucleons must a 3P_0 state and $T=1$. The cross-section near the threshold is given by $\sigma_{11} \sim 0.02 \eta^2$ ⁽⁸⁾ where η is the maximum pion momentum in the centre of mass system in units of meson mass.

Next consider the reaction (II)

$$\pi^+ + N + p \rightarrow N + N.$$

This corresponds to the inverse production reaction

$$p + p \rightarrow N + p + \pi^+$$

assuming charge symmetry in the production reaction. For the production reaction, the final nucleons are in the 3S_1 state and $T=0$, the meson being emitted in an S -state. The cross-section is given by $\sigma_{10} \sim 0.14\eta$. The contribution to the cross-section comes from the $T=0$ state for the final nucleons with initial protons (or neutrons) having $T=1$. We neglect the small 1S_0 contribution here. By applying detailed balance arguments one gets the following result

$$\frac{|\langle \pi^- N p | \mathcal{N} \mathcal{N} \rangle|}{|\langle \pi^- p p | \mathcal{N} p \rangle|} \approx \sqrt{\frac{\sigma_{10}}{\sigma_{11}}} \approx 2.65.$$

Here the η dependence is neglected as η and η^2 are small quantities of the same order. Also, the phase space factors cancel out, since the number of particles in the initial and final states are the same in the two basic reactions.

⁽⁸⁾ A. H. ROSENFELD: *Phys. Rev.*, **96**, 139 (1954).

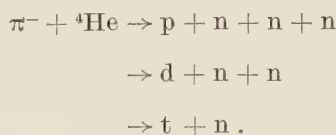
But, from our previous analysis we get

$$\frac{|\langle \pi \mathcal{N} p | \mathcal{N} \mathcal{N} \rangle|}{|\langle \pi p p | \mathcal{N} p \rangle|} \approx \frac{1}{\sqrt{2}} [1 + |b_1^{(0)}|^2]^{\frac{1}{2}} = 2.3.$$

Thus we see our solution checks rather well with the meson production cross-sections at threshold.

4. - Analysis of the π -meson absorption in helium and carbon.

A negative pion absorbed at rest in helium gives rise to the following reactions,



Where n, p, d, t are the neutron, proton, deuteron and triton respectively. We will show here that these reactions can be successfully explained by the foregoing analysis. Suppose the π^- -meson is captured by the (np)-pair in helium *i.e.*, $\pi^- + (np) + (np) \rightarrow (nn) + (np)$. After absorption, we have two fast neutrons, each having 55 MeV, going off in opposite directions while the other (np)-pair remaining as a spectator. The latter pair may come off as separate particle in which case the proton from this pair will have relatively low energy (~ 15 MeV). They may, however, come off as a deuteron but the possibility for the formation of deuteron can be shown to be very small. The deuteron formation may go through the following intermediate states: such as in the first stage one of the (np)-pair of helium participates in the absorption process while the other pair remaining as a spectator and finally the spectator (np)-pair may lead to the formation of a deuteron and a γ -ray through the n-p capture process. Since in the capture process the energy involved is of the order of MeV and as the cross-section of deuteron formation obeys the well known $(1/v)$ -law, we expect very little deuteron formation in the whole np-capture phenomena. In fact the matrix-element for such a process can be exhibited by

$$\left| \sum_n \frac{\langle \pi^- n p (np) | nn(np) \rangle \langle nn(np) | nn d \gamma \rangle}{E_i - E_n} \right|.$$

The cross-section for the process will evidently diminish the $1/v$ law by a factor of $1/\mu^2$ where μ is the meson mass and v is the relative velocity of the (n-p) pair available in the nucleus. Triton formation is even less likely, for,

then the neutron needed to make up the triton has much higher energy than the spectator (np)-pair.

If the capture takes place by a (pp)-pair (*i.e.*, $\pi^- + (pp) + (nn) \rightarrow (np) + (nn)$), we should get a pair of fast neutron and proton each with 55 MeV and going off in opposite directions, while the remaining two neutrons remaining as a spectator. Here the deuteron and triton formation are less likely for the same reason triton is in the previous case. So on the basis of our analysis we expect the relative ratio of slow to fast protons in helium to be the same as the ratio $\sigma(Np)/\sigma(pp)$ which, as we should expect from our analysis of Section 2, should be equal to 5. This is well borne out by the proton spectrum in helium as well as carbon (⁴).

RIASSUNTO (*)

Si presenta un'analisi dell'assorbimento dei π^- nei nuclei leggeri, supponendo che la reazione $\pi^- + (N^- \cdot N^0) \rightarrow N^- \cdot N^0$. Presupposte la indipendenza dell'interazione pione-nucleone dalla carica e la unitarietà e invarianza della matrice S all'inversione del tempo, si sono scritte in termini degli spostamenti di fase nucleone-nucleone le sezioni trasversali per le reazioni di cattura π^- con due nucleoni. I parametri che entrano nella teoria vengono determinati dalla osservazione sperimentale di OZAKI *et al.* Usando il principio dell'equilibrio dettagliato, si è fatto un controllo dei parametri con i dati di produzione dei mesoni alla soglia. Si mostra anche che il modello a due nucleoni fornisce una spiegazione coerente dei fenomeni di assorbimento del mesone π^- nell'elio e nel carbonio.

(*) Traduzione a cura della Redazione.

Anomalous Stark Effects in the Millimetre Wave Spectrum of Formyl Fluoride (*).

P. FAVERO (**) (***) and J. G. BAKER

Department of Physics, Duke University - Durham, North Carolina

(ricevuto il 6 Giugno 1960)

Summary. — Measurements of Stark effects in the rotational spectra of formyl and *d*-formyl fluoride show the presence of anomalous components that vary in strength with the field. They occur predominantly at high fields and may possibly be a molecular polarisability effect. In addition the normal molecular dipole moment is found to be $(1.99 \pm 0.03) D$, oriented almost parallel to the C-H bond and at an angle of 79° to the C-F bond. This is discussed in connection with the molecular structure.

1. — Introduction.

In a previous investigation of the rotational spectrum of formyl fluoride ⁽¹⁾ a curious doubling of some Stark components was noted. The present work studies this phenomenon in more detail and also yields a value for the molecular dipole moment from measurements of the shift of the Stark components at various fields. Measurements of this kind in the millimetre wave region have been restricted to light molecules ^(2,3,4) because Stark effects tend to fall off rapidly with increasing J ; however in the case of asymmetric top mole-

(*) This research was supported by the United States Air Force through the Air Force Office of Scientific Research under Contract AF 18(600)-497. Reproduction in whole or in part is permitted for any purpose of the United States Government.

(**) Present address: Istituto di Chimica Fisica, Università di Padova, Italy.

(***) International Fellow of the National Academy of Sciences through International Cooperation Administration.

(1) P. FAVERO, A. M. MIRRI and J. G. BAKER: *Journ. Chem. Phys.*, **31**, 556 (1959).

(2) R. TRAMBARULO, S. N. GHOSH, C. A. BURRUS and W. GORDY: *Journ. Chem. Phys.*, **21**, 851 (1953).

(3) C. A. BURRUS: *Journ. Chem. Phys.*, **28**, 427 (1958).

(4) B. BHATTACHARYA and W. GORDY: *Phys. Rev.*, in press; also D. M. LARKIN: private communication.

cules the decrease is relatively slow and is counterbalanced by the higher fields that may be obtained by using a smaller Stark cell.

2. - Experimental.

The spectrometer and 1 meter *K* band Stark cell used have been described elsewhere^(2,5). Observations were made with a 4000 Hz zero-based square wave voltage variable from 0 to 1000 V and measured by a peak to peak voltmeter. Lines and Stark components were detected by a lock-in amplifier and recorder or persistent screen oscilloscope (the latter by sweeping the klystron reflector with a 0.1 Hz saw-tooth wave form). The cell was calibrated by observations on the $0 \rightarrow 1$ transition of HCN at 88 632 MHz, for which very accurate measurements are available⁽⁴⁾. The strongest components were followed up to fields of 4000 V/cm and frequency shifts of 130 MHz before they were appreciably broadened by field inhomogeneities. However, the limiting accuracy was found to lie in reading the scale of the peak to peak voltmeter, which could not be done to better than 1%.

3. - Discussion.

The recorder tracings of Fig. 1 are typical of the patterns observed. It will be noted that doubling usually appears at fields of (1500÷2000) V/cm and the doublet splitting steadily increases with field. At the same time one component of the doublet remains constant in strength while the other increases rapidly. Frequency measurements have been made on all resolved Stark components of four transitions of HCOF

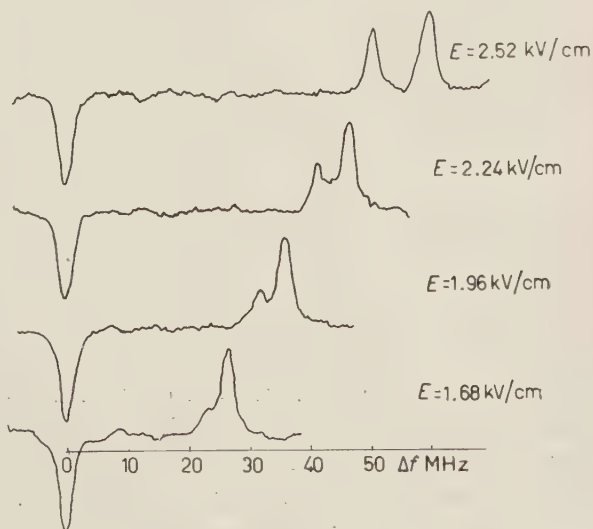


Fig. 1. - The $0_{00} \rightarrow 1_{11}$ transition of HCOF at 101 550.36 MHz. Stark components at various field strengths.

(5) P. FAVERO, A. M. MIRRI and J. G. BAKER: *Nuovo Cimento*, to be published.

and one transition of DCOF at fields ranging from 1000 to 4000 V/cm. The results are summarized in Table I which gives values of $\Delta f/E^2$ averaged over

TABLE I. - *Stark effect measurements.*

Transition	<i>M</i> value	$\Delta f/E^2$ MHz (kV/cm) ²	
		Observed	Calculated
$0_{00} \rightarrow 1_{11}$	0	9.4	9.37
	0''	8.3	
$1_{01} \rightarrow 2_{12}$	0	10.6 (broad) (unresolved)	9.30
	1		11.03
$5_{05} \rightarrow 5_{14}$	3	1.00	1.02
	3''	1.23	
	4	3.8 ₉	3.79
	4''	4.6 ₅	
	5	7.4 ₅	7.34
	5''	8.5	
$6_{06} \rightarrow 6_{15}$	5	4.2 ₅	4.21
	6	6.7	6.79
	6''	7.9 ₅	
$1_{01} \rightarrow 2_{12}$ (DCOF)	0	— 3.9 ₅	— 3.98
	1	+ 0.83	+ 0.81
	1''		

perceptible resolution
at 4000 V/cm.

Double primes denote anomalous components.

several measurements, with deviations up to 2% as expected from the volt-meter reading error. The normal components move linearly with E^2 as expected for an asymmetric top; the anomalous components (that is those varying in strength with field) do the same but with a markedly different slope. They also show some tendency to project back to a point that is not the unsplit line frequency, but experimental error is too large to give this tendency real significance. Fig. 2 shows a plot of the two components of the $0_{00} \rightarrow 1_{11}$ transition. These two are especially significant because this transition should have only one component ($M = 0$) and no perturbation would be expected to split this, as might happen for components of higher M .

Considerable care was taken to eliminate the possibility that this phenomenon was caused by an equipment defect. A two-valued field caused by

incorrect alignment of the Stark electrode does not account for the field dependence or the irregular slope of the anomalous components; furthermore no such effect was observed in the case of HCN although experimental conditions were the same. Irregularities in the square wave used would also have been noted for HCN. The field modulation was eliminated as a cause by carrying out video observations with a DC voltage taken from the rectifiers of the square wave generator; although the Stark components of the Table I transitions were a little too weak to measure, the doubling and intensity changes were clearly visible.

Calculations show that the anomalous components are not transitions of the form $\Delta M = \pm 1$, normally forbidden in the type of cell used here. A number of other cases of forbidden transitions produced by an electric field have been discussed (⁶) but none seem to correspond closely to the present work. The effect seems to be one of such high mole-

cular polarizability that an electric field can appreciably change the dipole moment. It is interesting to speculate on the uniqueness of this effect, for, although it has never been reported previously, work at fields of (2000–4000) V/cm has as yet been restricted to linear molecules. There is a considerable difference even between the two formyl fluoride isotopes, for in the DCOF transition of Table I the doubling only begins to appear at 4000 V/cm. DC field observations on the $8_{08} \rightarrow 8_{17}$ and $9_{09} \rightarrow 9_{18}$ transitions of this molecule similarly showed a doubling that just appeared at the highest available fields.

It seems that more accurate measurements at high fields on several molecules will be needed before this phenomenon of field dependent Stark doubling can be satisfactorily accounted for.

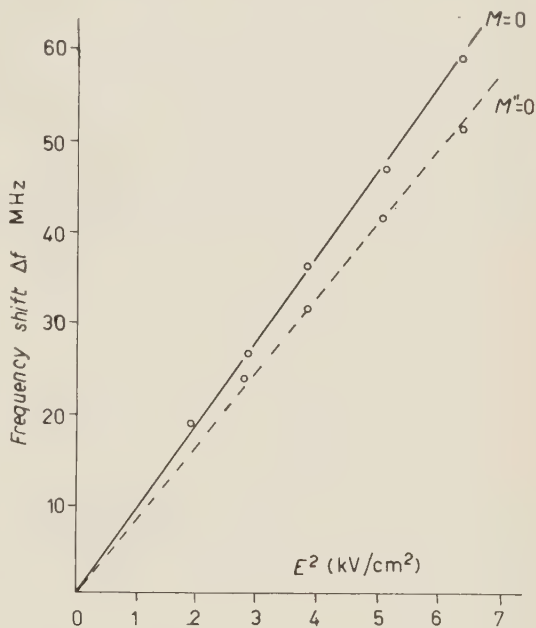


Fig. 2. — Stark displacements of $M=0$, $M''=0$ components of the $0_{00} \rightarrow 1_{11}$ transition.

(⁶) C. H. TOWNES and A. L. SCHAWLOW: *Microwave Spectroscopy* (New York, 1955), chap. 10.

4. - Dipole moment measurement.

Measurements on the normal Stark components have been fitted to the equation of Golden and Wilson (7):

$$\Delta W = (A + BM^2)E^2,$$

where A and B are complicated functions of the molecular transition probabilities and rotational energy differences. Although tables of the former are available (8) they are very widely spaced and it was found more satisfactory to calculate the required values from symmetric top transition probabilities and the asymmetric top transformation appropriate to the observed rotational constants (5). The dipole moment components of Table II fit the Table I

TABLE II. - Dipole moment components.

	HCOF	DCOF
μ_a	$(0.58 \pm 0.02) D$	$(0.61 \pm 0.02) D$
μ_b	$(1.91 \pm 0.03) D$	$(1.96 \pm 0.03) D$

measurements to approximately 2%. It is interesting that μ_b for DCOF is significantly larger than for HCOF. This effect appears in addition to the change due to a slight rotation of the principal axes of inertia. Although previous work (5) shows this rotation to be very slight ($\sim 1^\circ$) it does account for part of the change in the dipole moment components in going from HCOF to DCOF. Very accurate measurements would give a method of locating the position of the H atom, in the molecule, at present rather uncertain due to zero-point vibrations, by means of this rotation.

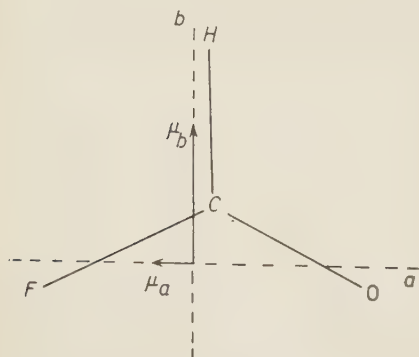


Fig. 3. - Principal inertial axes of HCOF.

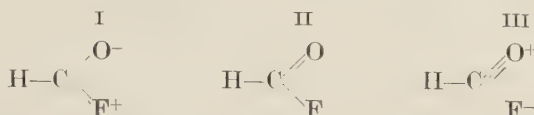
The change to DCOF will result in a rotation of the b axis towards the D atom. There is little doubt that μ_b is oriented with the H atom positive

(7) S. GOLDEN and E. B. WILSON: *Journ. Chem. Phys.*, **16**, 669 (1948).

(8) P. C. CROSS, R. M. HAINER and G. W. KING: *Journ. Chem. Phys.*, **12**, 210 (1944).

and the O and F atoms negative, and so for μ_a to increase on D substitution its orientation must be as shown. The total dipole moment of the molecule therefore has the value 1.99 ± 0.03 D and is oriented at 79° to the C-F bond and 13° to the C-H bond, with the hydrogen atom end positive.

Previously ⁽³⁾ three resonating structures were postulated on the basis of the observed bond lengths.



It now appears that the dipole moment corresponds more closely to structure I than to III, despite the bond lengths correspond better to III.

5. - Conclusion.

The existence of an anomalous doubling of the Stark components of formyl fluoride has been confirmed but there is no very satisfactory explanation for this. The normal molecular dipole moment is in excellent agreement with that determined by LEBLANC, LAURIE and GWINN ⁽⁹⁾ both in direction and orientation. These workers do not report any Stark doubling, but it would appear that they may not have reached sufficiently high fields with the Stark cell they described unless their square wave voltage was considerably higher than ours. Certainly much work remains to be done on this topic before it can be satisfactorily understood.

⁽⁹⁾ D. LEBLANC, V. W. LAURIE and W. D. GWINN: to be published.

Note added in proof.

D. M. LARKIN of the Physics Dept., Duke University, has examined the reported lines of HCOF further in a parallel plate Stark cell ⁽⁴⁾ and finds no doubling at fields up to 5000 V/cm. We feel that the arguments presented in the text are still valid, and it appears that if the extra Stark components are indeed spurious a new kind of equipment defect that merits much further investigation is occurring.

RIASSUNTO

La misura dell'effetto Stark nello spettro rotazionale del fluoruro di formile ha mostrato la presenza di componenti anomale che cambiano di intensità con il campo. Esse si presentano a campi elevati e possono essere dovute ad un effetto di polarizzazione molecolare. È stato determinato il momento dipolare normale il cui valore è (1.99 ± 0.03) D; esso è orientato quasi parallelamente al legame C-H e fa un angolo di 79° con il legame C-F. Questi dati sperimentali vengono discussi in connessione con la struttura molecolare.

Centrifugal Effects in Millimetre Wave Spectra: Formyl Fluoride (*).

P. FAVERO (**) (*), A. M. MIRRI (**) and J. G. BAKER

Department of Physics, Duke University - Durham, North Carolina

(ricevuto il 6 Giugno 1960)

Summary. — The three rotational and six distortion constants of both formyl and d-formyl fluoride have been determined from their millimetre wave absorption spectra, using an improved method of calculation of centrifugal effects. The rotational constants are: for HCOF, $A=91\,156.56$ MHz, $B=11\,760.23$ MHz, $C=10\,396.72$ MHz; for DCOF, $A=65\,096.59$ MHz, $B=11\,761.74$ MHz, $C=9\,941.71$ MHz. Taken with electron diffraction data these yield the parameters $r_{\text{CH}}=(1.087\pm0.01)$ Å, $r_{\text{CO}}=(1.182\pm0.003)$ Å, $r_{\text{CF}}=(1.341\pm0.003)$ Å, and $\beta_{\text{FCO}}=(123.04\pm0.02)^\circ$. Comparisons are made with the structure of the formyl grouping in similar molecules.

1. — Introduction.

Although rotational constants of molecules are now readily obtained by microwave spectroscopy to seven significant figures, this high precision does not carry over into the determination of internuclear distances and bond angles. The main cause of this is the presence of zero-point vibrations, which shift the molecular parameters from their equilibrium values by large and uncertain

(*) This work was supported by the United States Air Force through the Air Force Office of Scientific Research and Development Command. Reproduction in whole or in part is permitted for any purpose of the United States Government.

(**) Present address: Istituto di Chimica Fisica, Università di Padova, Italy.

(*) International Fellow of the National Academy of Sciences through International Cooperation Administration.

amounts. For example, KIVELSON, WILSON, and LIDE⁽¹⁾ studied a large number of isotopic forms of vinyl chloride but were not able to fix values of the bond lengths to better than 0.01 Å. Corrections for these vibration-rotation effects have been carried out for diatomic and some triatomic molecules, but the problem has become excessively difficult in more complicated cases. However little attention has been paid to centrifugal effects, although these depend on the molecular vibrations in a markedly similar manner.

In addition to the fundamental difficulty above, it is often necessary to assume values for some molecular bond lengths because of the lack of sufficient data to determine them all. These values are chosen by comparison with the parameters in similar molecules for which a full determination exists. A particularly common practice is to take a C-H bond length of 1.11 Å, as determined for the methyl halides⁽²⁾, but recently COSTAIN⁽³⁾ has pointed out that studies by isotopic substitution in these halides support a bond length of only 1.096 Å. Another recent change in an accepted bond length is shown by the C=C bond in ethylene, which formerly was given by an infrared study⁽⁴⁾ as 1.353 Å. Three new determinations, by electron diffraction⁽⁵⁾, infrared spectroscopy⁽⁶⁾, and Raman spectroscopy⁽⁷⁾, agree in setting this length in the vicinity of 1.335 Å. It would seem that many of the rotational analyses of complicated molecules are based on data that are, at best, uncertain, and there is great need of a detailed analysis of the behaviour of simple chemical groupings in changing molecular surrounding.

The formyl grouping, HCO—, is particularly well adapted to such a study. Detailed measurements are available for formaldehyde⁽⁸⁾, formic acid^(9,10), methyl formate⁽¹¹⁾, formamide⁽¹²⁾, and acetaldehyde⁽¹³⁾. However, in several of these molecules there are further complications due to internal rotation. Formyl fluoride, for which no microwave studies have previously appeared in

(1) D. KIVELSON, E. B. WILSON and D. R. LIDE: *Journ. Chem. Phys.*, **32**, 205 (1960).

(2) S. L. MILLER, L. C. AAMODT, G. DOUSMANIS, C. H. TOWNES and J. KRAITCHMAN: *Journ. Chem. Phys.*, **20**, 1112 (1952).

(3) C. C. COSTAIN: *Journ. Chem. Phys.*, **29**, 864 (1958).

(4) W. S. GALLAWAY and E. F. BARKER: *Journ. Chem. Phys.*, **10**, 88 (1942).

(5) L. S. BAPTELL and R. A. BONHAM: *Journ. Chem. Phys.*, **27**, 1414 (1957).

(6) H. C. ALLEN and E. K. PLYLER: *Journ. Amer. Chem. Soc.*, **80**, 2673 (1958).

(7) J. M. DOWLING and B. P. STOICHEFF: *Ann. Rev. Phys. Chem.*, **10**, 33 (1959).

(8) R. B. LAWRENCE and M. W. P. STRANDBERG: *Phys. Rev.*, **83**, 363 (1951).

(9) R. TRAMBARULO, A. CLARK and C. HEARNS: *Journ. Chem. Phys.*, **28**, 736 (L) (1958).

(10) G. LERNER, B. P. DAILEY and J. P. FRIEND: *Journ. Chem. Phys.*, **26**, 680 (1957); also A. M. MIRRI, to be published.

(11) R. F. CURL: *Journ. Chem. Phys.*, **30**, 1529 (1959).

(12) C. C. COSTAIN and J. M. DOWLING: *Journ. Chem. Phys.*, **32**, 158 (1960).

(13) R. W. KILB, C. C. LIN and E. B. WILSON: *Journ. Chem. Phys.*, **26**, 1695 (1957).

the literature, is conveniently simple and possesses the advantage of having a dipole moment not orientated along any principal axis, so that it is possible to observe rotational transitions originating in low energy states to determine the rotational constants and in high energy states to measure centrifugal effects. Both types of transition can be observed in the 2-3 mm wavelength region.

2. - Experimental.

Formyl fluoride was made from formic acid, KF and benzoyl chloride, as described by MORGAN, STAATS and GOLDSTEIN ⁽¹⁴⁾. It was found that even traces of water greatly lowered the yield, and rigorous drying of the reagents, by means of anhydrous Cu SO_4 for the liquids and strong heating for the KF, was necessary to bring the yield to the 20 % quoted by the above authors. d-Formyl fluoride, DCOF , was prepared in the same way from a 5 ml sample of 97 % DCOOD , sent to us by Dr. H. W. MORGAN of Oak Ridge National Laboratory, to whom we are deeply indebted. The gas, with boiling point -26°C , was collected at dry ice temperature and stored under liquid nitrogen after distillation with retention of the middle fraction. All microwave studies were carried out at dry ice temperature to avoid decomposition to CO and HF at room temperature: even at dry ice temperature this change becomes noticeable after some weeks.

Millimetre waves were generated by using a harmonic generator of the type described by KING and GORDY ⁽¹⁵⁾ to multiply up the output of a 2K33 K band klystron. The output was fed from fourth harmonic waveguide ((0.075 ± 0.034) in.) to a two metre K band absorption cell with mica windows and detected by a fourth harmonic or sixth harmonic (guide dimensions: (0.045 ± 0.022) in.) detector. Lines were observed on a cathode ray oscilloscope by video detection; Stark patterns were studied by means of a conventional one metre K band Stark cell, using 4000 Hz field modulation and a lock-in amplifier. Frequency measurements were made at the fundamental frequency by means of a frequency standard monitored by Station WWV, whose input at 5 MHz was correct to a few parts in 10^9 . The limiting accuracy of frequency measurement was set by the rather large line width and is estimated as ± 0.04 MHz at the fundamental frequency. Final identification of the correct harmonic at which a line appeared was made by tuning this line with a stub at the detector and finding its wavelength in the guide from the

⁽¹⁴⁾ H. W. MORGAN, P. A. STAATS and J. H. GOLDSTEIN: *Journ. Chem. Phys.*, **25**, 337 (1956).

⁽¹⁵⁾ W. C. KING and W. GORDY: *Phys. Rev.*, **93**, 407 (1954).

separation of its successive disappearances. It was readily possible to distinguish between the first three harmonics transmitted by the detector guide in this way, but more difficult for higher harmonics.

3. - Analysis of spectra.

Although neither gas showed lines with a resolvable hyperfine structure, a very large number of lines without any regular pattern occurred due to the presence of *b*-type transitions of much greater intensity than *a*-type transitions. The method of analysis was to search for *Q* branch transitions, which were readily identifiable by their strength and their characteristic Stark pattern. When sufficient of these had been found it was possible to compare their frequencies with those predicted from the crude molecular model used by MORGAN, STAATS, and GOLDSTEIN ⁽¹⁾ and to modify the rotational constants to get better agreement; only the assignments in Tables I and II gave close agreement.

TABLE I. - *Transitions of HCOF.*

Assignment	Frequency Observed	Δf Calc. - Obs.	Centrifugal Shift
<i>a</i> -type transitions			
$6_{06} \rightarrow 7_{07}$	154 120.02 MHz	+ 0.45 MHz	- 12.37 MHz
$6_{16} \rightarrow 7_{17}$	150 098.73	- 0.99	- 8.78
$6_{15} \rightarrow 7_{16}$	159 618.91 (*)	- 0.02	- 14.58
$6_{24} \rightarrow 7_{25}$	155 923.97	+ 3.66	- 8.59
$6_{25} \rightarrow 7_{26}$	154 955.55	+ 0.55	- 7.08
$6_{33} \rightarrow 7_{34}$	155 251.93	- 0.08	- 0.30
$6_{34} \rightarrow 7_{35}$	155 234.17	+ 0.26	+ 0.24
$6_4 \rightarrow 7_4$	155 195.15	+ 0.14	+ 10.03
$6_5 \rightarrow 7_5$	155 183.56	+ 0.10	+ 23.24
$6_6 \rightarrow 7_6$	155 186.37	+ 0.34	+ 39.37
<i>b</i> -type transitions			
$0_{00} \rightarrow 1_{11}$	101 550.36 MHz	- 0.05 MHz	- 2.97 MHz
$1_{01} \rightarrow 2_{12}$	122 343.95	- 0.04	- 2.75
$2_{02} \rightarrow 3_{13}$	142 462.44	- 0.07	- 2.63
$3_{03} \rightarrow 4_{14}$	161 927.56	- 0.12	- 2.68
$5_{14} \rightarrow 4_{23}$	119 298.55	+ 0.07	- 34.93
$6_{15} \rightarrow 5_{24}$	93 164.04	- 0.04	- 26.52
$8_{18} \rightarrow 9_{09}$	138 988.38	+ 0.13	- 37.07
$5_{05} \rightarrow 5_{14}$	90 786.64	+ 0.17	- 2.09
$6_{06} \rightarrow 6_{15}$	95 339.16	+ 0.37	- 2.99
$7_{07} \rightarrow 7_{16}$	100 838.05	+ 0.13	- 5.13
$8_{08} \rightarrow 8_{17}$	107 362.11	- 0.63	- 9.13
$9_{09} \rightarrow 9_{18}$	114 993.80	- 1.54	- 15.77
$10_{0,10} \rightarrow 10_{19}$	123 812.35	+ 0.08	- 25.96

(*) Calculated by combining other transition frequencies.

TABLE II. — *Transitions of DCOF.*

Assignment	Frequency Observed	Δf Calc. — Obs.	Centrifugal Shift
<i>a</i> -type transitions			
$6_{06} \rightarrow 7_{07}$	149 439.56 MHz	— 0.33 MHz	— 10.56 MHz
$6_{16} \rightarrow 7_{17}$	144 988.68	— 0.15	— 8.53
$6_{15} \rightarrow 7_{16}$	157 633.14	— 0.02	— 13.89
$6_{24} \rightarrow 7_{25}$	154 029.84	0.00	— 11.56
$6_{25} \rightarrow 7_{26}$	151 558.74	— 0.03	— 9.63
$6_{33} \rightarrow 7_{34}$	152 343.06	+ 0.16	— 7.59
$6_{34} \rightarrow 7_{35}$	152 253.06	— 0.06	— 7.55
$6_{42} \rightarrow 7_{43}$	152 147.81	+ 1.15	— 3.73
$6_{43} \rightarrow 7_{44}$	152 147.81	+ 0.06	— 3.72
$6_5 \rightarrow 7_5$	152 088.00	— 0.24	+ 1.13
$6_6 \rightarrow 7_6$	152 059.08	+ 0.29	+ 7.05
<i>b</i> -type transitions			
$1_{01} \rightarrow 2_{12}$	94 920.60 MHz	— 0.09 MHz	— 1.19 MHz
$2_{02} \rightarrow 3_{13}$	113 911.30	— 0.04	— 1.41
$3_{03} \rightarrow 4_{14}$	132 067.57	+ 0.27	— 1.91
$4_{04} \rightarrow 5_{15}$	149 483.49	+ 0.21	— 3.22
$3_{12} \rightarrow 2_{21}$	92 176.92	— 0.16	— 16.41
$6_{25} \rightarrow 5_{32}$	141 567.45	+ 0.08	— 67.25
$2_{12} \rightarrow 2_{21}$	165 448.32	— 1.17	— 17.49
$3_{13} \rightarrow 3_{22}$	168 210.00	— 5.00	— 17.95
$3_{12} \rightarrow 3_{21}$	157 515.96	— 0.14	— 16.48
$4_{13} \rightarrow 4_{22}$	154 391.16	+ 0.19	— 15.19
$5_{14} \rightarrow 5_{23}$	150 856.20	+ 0.15	— 13.43
$6_{15} \rightarrow 6_{24}$	147 150.05	+ 0.43	— 11.29
$7_{16} \rightarrow 7_{25}$	143 547.24	— 0.05	— 8.95
$8_{17} \rightarrow 8_{26}$	140 340.46	+ 0.07	— 6.82
$8_{08} \rightarrow 8_{17}$	93 881.72	+ 0.19	— 12.89
$9_{09} \rightarrow 9_{18}$	105 584.79	+ 0.27	— 21.50
$10_{0,10} \rightarrow 10_{19}$	119 163.65	+ 0.21	— 33.94
$11_{0,11} \rightarrow 11_{1,10}$	134 614.08	— 0.12	— 50.96
$12_{0,12} \rightarrow 12_{1,11}$	151 855.87	— 0.09	— 73.04

These *Q* branch frequencies depend on only two rotational constants, but the third was estimated by assuming planarity of the molecule and a search carried out for *R* branch transitions of low *J* in the vicinity of the predicted frequencies. These transitions were also identified by their Stark patterns, which consist of only a few components. The best rotational constants without taking account of centrifugal distortion were derived from the low *J* transitions and used to locate transitions needed to complete the centrifugal distortion fitting.

Rotational transition frequencies were fitted to the expression of King,

Hainer, and Cross:

$$W/h = \frac{1}{2}(A + C)J(J+1) + \frac{1}{2}(A - C)E_{\tau}$$

using published tables of E_{τ} . These are described in reference (16). For centrifugal effects a convenient new formula (17) was devised, based on the molecular «reduced energy» w and asymmetry parameter b , also described in reference (16).

$$\begin{aligned} W'/h = & b R_{10} J(J+1) - \frac{5}{2} b w R_{10} - (D_J + \frac{1}{2} b R_{10}) J^2(J+1)^2 - \\ & - (D_{JK} - b R_{10}) J(J+1)(w - b dw/db) - 2\delta_J J(J+1) dw/db + \\ & + R_{10} w dw/db + (R_6 - \frac{1}{4} b R_{10}) T_{JK}. \end{aligned}$$

Here R_{10} has the value $(b D_K + 4 R_5)/(1 - \frac{1}{2} b^2)$ in terms of constants defined by NIELSEN (18). The correction term T_{JK} is only important in the following cases:

$$K = 0 \quad T_{JK} = \frac{1}{16} b^2 (J-3)(J-2)(J-1) J(J+1)(J+2)(J+3)(J+4),$$

$$K = 1 \quad T_{JK} = \frac{8c_1 R_5}{b} (w - 1 - c_1 b),$$

$$K = 2 \quad (E^{\pm}) \quad T_{JK} = \pm (J-1) J(J+1)(J+2).$$

All these coefficients may be calculated from the tables of E_{τ} by means of suitable interpolation and the algebraic relations between w and E_{τ} . The calculations were programmed onto the Mathematics Department IBM 650 computer, with a resultant great saving of time. Initial values of the constants were obtained by Erlandsson's method (19) from the a -type transitions and were then refined by iteration. The transition frequencies were rather insensitive to R_5 and R_6 which were instead estimated from the following relations, true for any molecule with one plane of symmetry:

$$\begin{aligned} R_5 = & \frac{1}{2}(D_J + \delta_J - \frac{1}{2} D_{JK}) - I_b^2/2I_a^2 \times (D_J + 6R_6) - \\ & - I_a^2/I_c^2 \times (D_J + D_{JK} + D_K) + I_b^4/2I_a^2 I_c^2 \times (D_J - 2\delta_J - 2R_6), \end{aligned}$$

$$R_6 = \frac{[(I_c^2 - I_b^2)^2 - I_a^4] D_J - I_a^4 (D_{JK} + D_K) + 2(I_c^4 - I_b^4) \delta_J}{2(I_c^4 + 6I_c^2 I_b^2 + I_b^4)}.$$

(16) C. H. TOWNES and A. L. SCHAWLOW: *Microwave Spectroscopy* (New York, 1955).

(17) J. G. BAKER: *Bull. Am. Phys. Soc.*, **5**, 241 (1960).

(18) H. H. NIELSEN: *Rev. Mod. Phys.*, **23**, 213 (1951).

(19) G. ERLANDSSON: *Journ. Chem. Phys.*, **28**, 71 (1958).

The constants obtained are shown in Table III. A most significant point is the very large value of D_K , which contributes even to the low J transitions. Neglect of this leads to an error in A of the order of D_K , and so it appears that determinations of the largest rotational constants of near-prolate rotors are not to be trusted unless corrected for centrifugal effects. Another interesting fact is the almost threefold change in D_{JK} between the two isotopes; this is probably associated with the closeness of the light H atom to the b axis of inertia.

TABLE III. — *Constants of formyl fluoride.*

	HCOF	DCOF
A MHz	91 156.56	65 096.59
B MHz	11 760.23	11 761.74
C MHz	10 396.72	9 941.71
κ	— 0.966 233 0	— 0.934 002 9
D_J kHz	9.85	8.82
D_{JK} kHz	— 105.5	— 38.1
D_K kHz	3 150	1 207
δ_J kHz	— 2.13 ₅	— 2.00
R_5 kHz	25	4
R_6 kHz	— 0.135	— 0.162
R_{10} kHz	73	— 4
I_a a.m.u. \AA^2	5.545 7 ₅	7.765 8 ₅
I_b a.m.u. \AA^2	42.986 5	42.981 0
$I_c - I_a - I_b$ a.m.u. \AA^2	0.091 8 ₅	0.1027

It will be seen that the lines of Tables I and II have been fitted within the experimental error of 0.04 MHz at 25 kMHz, except for a few isolated and unexplained cases, and it appears that the formula given accounts rather well for centrifugal effects in an asymmetric molecule.

4. — Molecular structure.

It is possible to locate the H atom in the centre-of-gravity co-ordinates of HCOF by using Kraitchman's method of isotopic substitution⁽²⁰⁾. COSTAIN⁽³⁾ recommends the use of the two in-plane moments of inertia so long as the inertial defects of the two (planar) molecules are not too different. However, use of I_b leads to an imaginary co-ordinate because the H atom is very close

(20) J. KRAITCHMAN: *Amer. Journ. Phys.*, **21**, 17 (1953).

to the b axis and zero-point vibrations make this quantity smaller for DCOF than for HCOF. This problem does not arise if I_a and I_c are treated as the equilibrium moments of inertia, as will be done here. The result is shown in Table IV.

TABLE IV. — *Centre-of-gravity coordinates in HCOF.*

	H	C	O	F
$b(A)$	1.5007	0.4150	— 0.1749	— 0.1945
$a(A)$	0.0713	0.1325	1.1573	— 1.0620
$r_{\text{CH}}(1.087 \pm 0.01) \text{ \AA}$			$\beta_{\text{FCO}}(123.04 \pm 0.02)^\circ$	
$r_{\text{CO}}(1.182 \pm 0.003) \text{ \AA}$			$\beta_{\text{FCH}} 114^\circ$	
$r_{\text{CF}}(1.341 \pm 0.003) \text{ \AA}$			$\beta_{\text{OCH}} 123^\circ$	

Even though the H atom is definitely located there remain only two moments of inertia to locate the remaining three atoms. It is necessary to use the most accurate electron diffraction data ⁽²¹⁾ ($r_{\text{CO}} = (1.192 \pm 0.011) \text{ \AA}$; $r_{\text{CF}} = (1.351 \pm 0.013) \text{ \AA}$; $r_{\text{CF}}/r_{\text{CO}} = (1.134 \pm 0.005) \text{ \AA}$) to choose a final structure. Fortunately these restrictions are sufficient to locate the other three atoms with considerable accuracy; the best centre-of-gravity co-ordinates and a final structure are given in Table IV. This agrees very well with the results obtained by LEBLANC, LAURIE, and GWINN ⁽²²⁾ from studies at lower frequencies. It should be emphasized that the error limits given do not take account of zero-point vibration corrections; these will probably not affect the bond lengths significantly but will produce an uncertainty of several degrees in the angles

TABLE V. — *The formyl group in some simple molecules.*

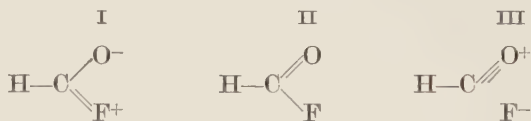
	C — H	C = O
HCOF	1.087 \AA	1.182 \AA
H ₂ CO ⁽⁸⁾	1.12	1.21
HCOOH ^(9,10)	1.092	1.237
HCONH ₂ ⁽¹²⁾	1.102	1.19 ₃
HCOOCH ₃ ⁽¹¹⁾	1.10	1.20

involving the C—H bond. One of us (J.G.B.) has devised a method of using centrifugal constants to estimate these corrections, details of which will be

⁽²¹⁾ M. E. JONES, K. HEDBERG and V. SCHOMAKER: *Journ. Amer. Chem. Soc.*, **77**, 5278 (1955).

⁽²²⁾ D. LEBLANC, V. W. LAURIE and W. D. GWINN: to be published.

published elsewhere, but further isotopic studies are required before they can be made for formyl fluoride. It is safe to conclude that the formyl fluoride molecule is exceptionally compact, for the C=O length is shorter than in all the molecules of Table V while the C—F bond is much shorter than the single bond length (1.385 Å) found in CH₃F⁽²³⁾. It appears that there are significant contributions from all three structures below:



5. — Conclusion.

A rotational and centrifugal analysis of the millimetre wave absorption spectra of HCOF and DCOF has led to a determination of all nine constants involved for each molecule and the conclusion that it is not safe to neglect centrifugal effects even for low J transitions. The molecular parameters obtained are in good agreement with those of earlier workers, and show that the formyl grouping undergoes large changes even in the simplest formyl compounds.

* * *

The authors would like to express their thanks to Dr. T. M. GALLIE for his help in setting up the computer programme and to Professor WALTER GORDY for his sustained interest throughout this project.

(²³) O. R. GILLIAM, H. D. EDWARDS and W. GORDY: *Phys. Rev.*, **75**, 1014 (1949).

RIASSUNTO

Sono state determinate le tre costanti rotazionali e le sei costanti di distorsione centrifuga del fluoruro di formile e del deutero fluoruro di formile studiandone gli spettri di assorbimento nella regione millimetrica e usando un metodo perfezionato di calcolo degli effetti centrifughi. Le costanti rotazionali sono: per HCOF, $A=91156.56$ MHz, $B=11760.23$ MHz, $C=10396.72$ MHz; per DCOF, $A=65096.59$ MHz, $B=11761.74$ MHz, $C=9941.71$ MHz. Tenendo conto dei dati di diffrazione elettronica, queste costanti rotazionali forniscono i seguenti valori per i parametri molecolari: $r_{\text{OH}}=1.087 \pm 0.01$ Å, $r_{\text{CO}}=(1.182 \pm 0.003)$ Å, $r_{\text{CF}}=(1.341 \pm 0.003)$ Å e $\beta_{\text{FCO}}=(123.04 \pm 0.02)^\circ$.

Test of Global Symmetry from Multinucleon K^- -Capture.

S. S. SAXENA

Theoretical Physics and Applied Mathematics Divisions, A.E.E.T. - Bombay

S. N. BISWAS

Tata Institute of Fundamental Research - Bombay

(ricevuto il 6 Giugno 1960)

Summary. — The principle of charge-independence coupled with the unitarity and time-reversibility of the scattering matrix enables one to express the production amplitude of the K^- -capture reactions in terms of the scattering phases of the final state interaction. By making impulse type approximation, certain relations between $K^- + d$ and $K^- + 2N$ absorption cross-sections are obtained. The experimental data have been analysed on the basis of the global symmetry of strong interactions. It is found that *within* the limits of our approximation this symmetry is incompatible with the experimental observations.

1. — Introduction.

Serious doubts have already been cast on the usefulness of the concept of the global symmetry of pion-baryon interaction. SALAM⁽¹⁾ and others^(2,4) by analysing the K^- -meson-nucleon scattering and absorption data have re-

(¹) A. SALAM: *Report on Kiev Conference on High Energy Physics* (1959). Also his Lecture at 47-th Indian Science Congress (Bombay, 1960).

(²) M. ROSS and G. SHAW: *Phys. Rev.*, **115**, 1773 (1959).

(³) B. D'ESPAGNAT and J. PRENTKI: *Nuovo Cimento*, **15**, 130 (1960).

(⁴) T. SAKUMA and S. FURUI: *Prog. Theor. Phys.*, **23**, 522 (1960).

jected the global symmetry. Recently it has been observed⁽⁵⁾ that about 35% of all K^- -captures at rest in heavy nuclei proceeds via the multinucleon mode. This observation is based upon the identification of fast baryons which are emitted only if the K^- was absorbed by two nucleons simultaneously leading to the reactions



An estimate of the various reaction rates in (1) is now available⁽⁵⁾.

We propose here to examine the compatibility of the global symmetry with these experimental observation of multinucleon K^- capture. As a method of testing we have, by the use⁽⁶⁾ of time-reversibility and unitarity of the S -matrix coupled with charge independence, explicitly expressed the cross-section of hyperon production in (1) in terms of the hyperon-nucleon scattering phase-shifts. Since by the hypothesis of global symmetry the hyperon-nucleon interaction may very well be represented by the nucleon-nucleon interaction hence the analysis of the reaction cross-sections with the observed branching ratios may predict the hyperon-nucleon scattering phase shift which is easily checked with the observed nucleon-nucleon phase shift at low energy. It should be mentioned that the experimental situation regarding multinucleon K^- -capture is not very clear; however, a good insight⁽⁵⁾ of the situation can be had if one can establish some relations among the $K^- + 2N$ and $K^- + d$ reaction cross-sections. For this one makes the following impulse type approximation. Since deuteron is a loosely bound system one can assume that reaction proceeds as if the K^- interacts directly with the nucleons being predominantly in the iso-singlet state. It is seen that under this approximation there exists certain equality among the cross-section of $K^- + 2N$ and $K^- + d$ absorption reactions. Since we confine our analysis of the data to low energy, K^- is assumed to be absorbed in S -state only, but no definite conclusion is yet available.

2. - Isotopic spin analysis.

By the global symmetry hypothesis the isotopic doublet

$$(2) \quad \left\{ \begin{array}{l} Y = (\Sigma^+, Y^0) \quad \text{or} \quad Z = (Z^0, \Sigma^-) \\ \text{with} \\ A^0 = \frac{1}{\sqrt{2}} (Z^0 + Y^0) \quad \text{and} \quad \Sigma^0 = \frac{1}{\sqrt{2}} (Z^0 - Y^0), \end{array} \right.$$

⁽⁵⁾ Y. EISENBERG, W. KOCH, M. NIKOLIĆ, M. SCHNEEBERGER and H. WINZELER: *Nuovo Cimento*, **11**, 351 (1959), also. European Collaboration Data: reported by E. H. S. BURHOP at the Kiev Conference (1959); Y. EISENBERG and W. KOCH: *Nuovo Cimento*, **11**, 453 (1959).

⁽⁶⁾ K. M. WATSON: *Phys. Rev.*, **95**, 228 (1954). See also: E. FERMI: *Suppl. Nuovo Cimento*, **2**, 59 (1955) and B. TOUSCHKE: *Suppl. Nuovo Cimento* **14**, 278 (1959).

is coupled to the pion in the absence of K -meson interaction, in identically the same way as the nucleon doublet, \mathcal{N} . We note that the \mathcal{N} - Y or \mathcal{N} - Z system is then in the isotopic spin states $I = 1$ and 0 . Correspondingly we can then define two amplitudes a_1 and a_0 in terms of which all the \mathcal{N} - Y or \mathcal{N} - Z scattering can be expressed.

On the other hand, the \mathcal{N} - Σ system is characterized in general, with the usual Gell-Mann-Nishijima assignment of isospin, by the states $\Phi_{\frac{3}{2}}$ and $\Phi_{\frac{1}{2}}$ corresponding to the isospin states $T = \frac{3}{2}$ and $\frac{1}{2}$ respectively and the \mathcal{N} - Λ system by the state $\Psi_{\frac{1}{2}}$ of $T = \frac{1}{2}$ only.

If S_0 denotes the scattering matrix in the absence of K -interaction then one can easily express $\mathcal{N} - \Sigma$, $\mathcal{N} - \Lambda$ scattering amplitudes in terms of a_1 and a_0 . We have thus

$$(3) \quad \left\{ \begin{array}{l} \langle \Phi_{\frac{3}{2}} | S_0 | \Phi_{\frac{3}{2}} \rangle = a_1 ; \quad \langle \Phi_{\frac{1}{2}} | S_0 | \Phi_{\frac{1}{2}} \rangle = \frac{1}{4} (3a_0 + a_1) , \\ \langle \Psi_{\frac{1}{2}} | S_0 | \Psi_{\frac{1}{2}} \rangle = \frac{1}{4} (3a_1 + a_0) \quad \text{and} \quad \langle \Psi_{\frac{1}{2}} | S_0 | \Phi_{\frac{1}{2}} \rangle = \frac{\sqrt{3}}{4} (a_1 - a_0) . \end{array} \right.$$

We note from (3) that S_0 has non-vanishing matrix elements between $\Phi_{\frac{1}{2}}$ and $\Psi_{\frac{1}{2}}$ states and hence it is not diagonal. To diagonalize the S_0 -matrix we superimpose the $\Phi_{\frac{1}{2}}$ and $\Psi_{\frac{1}{2}}$ states so as to obtain the states $\eta_{\frac{1}{2}}$ and $\varepsilon_{\frac{1}{2}}$: We write

$$(4) \quad \left\{ \begin{array}{l} \eta_{\frac{1}{2}} = \frac{1}{2} \Phi_{\frac{1}{2}} + \frac{\sqrt{3}}{2} \Psi_{\frac{1}{2}} , \\ \varepsilon_{\frac{1}{2}} = -\frac{\sqrt{3}}{2} \Phi_{\frac{1}{2}} + \frac{1}{2} \Psi_{\frac{1}{2}} . \end{array} \right.$$

We have then from (3) ⁽⁷⁾

$$(5a) \quad \langle \eta_{\frac{1}{2}} | S_0 | \eta_{\frac{1}{2}} \rangle = a_1$$

$$(5b) \quad \langle \varepsilon_{\frac{1}{2}} | S_0 | \varepsilon_{\frac{1}{2}} \rangle = a_0$$

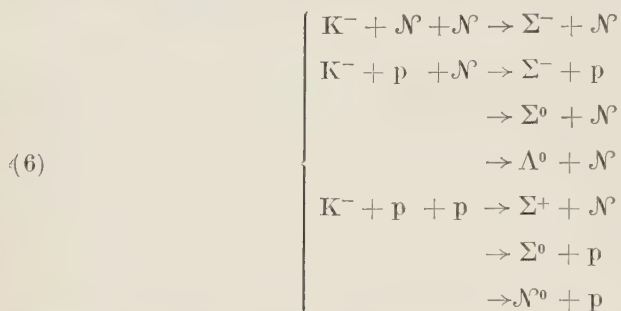
and

$$(5c) \quad \langle \eta_{\frac{1}{2}} | S_0 | \varepsilon_{\frac{1}{2}} \rangle = 0 .$$

⁽⁷⁾ Cf. AMATI and VITALE: *Fort. der Physik*, **7**, 416, (1959).

3. - Absorption cross-sections and analysis of the data.

The above analysis would now enable us to obtain⁽⁸⁾ the following absorption processes incorporating the global symmetry hypothesis. Consider the reactions:



The initial system can be described by three states $\chi_{\frac{3}{2}}^{(1)}$, $\chi_{\frac{1}{2}}^{(1)}$ and $\chi_{\frac{1}{2}}^{(0)}$ where the lower suffix denotes the total isotopic spin and the upper index denotes the total isotopic spin of the initial two nucleons. We have already seen that the final (ΣN) and (ΛN) system could be expressed in terms of the states $\eta_{\frac{1}{2}}$ and $\varepsilon_{\frac{1}{2}}$, by the principle of conservation of isotopic spin all the reaction in (6) can be expressed by the following S -matrix elements:

$$(7) \quad \left\{ \begin{array}{l} \langle \chi_{\frac{3}{2}}^{(1)} | S | \Phi_{\frac{3}{2}} \rangle = D \\ \langle \chi_{\frac{1}{2}}^{(1)} | S | \eta_{\frac{1}{2}}^{(1)} \rangle = B_1 \\ \langle \chi_{\frac{1}{2}}^{(1)} | S | \varepsilon_{\frac{1}{2}} \rangle = C_1 \end{array} \right. \quad \left\{ \begin{array}{l} \langle \chi_{\frac{1}{2}}^{(0)} | S | \eta_{\frac{1}{2}} \rangle = B_0 \\ \langle \chi_{\frac{1}{2}}^{(0)} | S | \varepsilon_{\frac{1}{2}} \rangle = C_0, \end{array} \right.$$

where the B 's, C 's and D are all complex quantities.

In accordance with our assumption already mentioned in the introduction about the mechanism of $K^- + d$ reaction we can also represent this $(K^- + d)$ system by $\chi_{\frac{1}{2}}^{(0)}$ state. Hence with the help of the above defined quantities it is now an easy matter to obtain the transition matrix-elements for the process of K^- absorption by deuteron, namely

$$(8) \quad \left\{ \begin{array}{l} K^- + d \rightarrow \Sigma^- + p \\ \qquad \qquad \qquad \rightarrow \Sigma^0 + N \\ \qquad \qquad \qquad \rightarrow \Lambda^0 + N. \end{array} \right.$$

Now by decomposing the pure physical state of interest in terms of the eigenstates of the total isotopic-spin operator T , we can obtain all the transition amplitudes for the reactions in (6) and (8) in terms of the B 's, C 's and D only.

⁽⁸⁾ For details of the calculation see: Y. EISENBERG and W. KOCII: *Nuovo Cimento*, **11**, 453 (1959).

Denote the phase shifts for the hyperon-nucleon scattering by α_1 and α_0 corresponding to the total isospin state 1 and 0 (in global symmetry) (α 's are real). From the unitarity and time-reversal invariance of the S -matrix (*) we at once write

$$(8^*) \quad \begin{cases} D = |D| \exp [i\alpha_1], & B_1 = |B_1| \exp [i\alpha_1], & C_1 = |C_1| \exp [i\alpha_1] \\ B_0 = |B_0| \exp [i\alpha_0], & C_0 = |C_0| \exp [i\alpha_0], \end{cases}$$

where $|D|$, $|B|$'s, $|C|$'s are all real and positive quantities and equations (8*) are defined within a common irrelevant phase factor.

From the experimental situation (5) concerning the multi-nucleon reaction rates it seems that the frequency of the $K^- + 2N$ capture leading to $\Sigma^- + N$ is very small compared with other production yields. The cross-section $\sigma(K^- + N N \rightarrow \Sigma^- N)$ using (7) and (8*) is proportional to $|D|^2$ hence from the experimental evidence one sets $|D| = 0$. Using this and from (7) and (8*) we have the following result:

$$(9) \quad \left\{ \begin{aligned} \sigma(K^- + pN \rightarrow \Sigma^- + p) &\approx \frac{1}{36} |B_1|^2 + \frac{1}{12} |C_1|^2 + \frac{1}{12} |B_0|^2 + \frac{1}{4} |C_0|^2 - \\ &\quad - \frac{1}{6\sqrt{3}} |B_1| |C_1| \cos \delta - \frac{1}{2\sqrt{3}} |B_0| |C_0| \cos \delta, \\ \sigma(K^- + pN \rightarrow \Sigma^0 + N) &\approx \frac{1}{2} \sigma(K^- + pN \rightarrow \Sigma^- + p), \\ \sigma(K^- + pN \rightarrow \Lambda^0 + N) &\approx \frac{1}{8} |B_1|^2 + \frac{1}{24} |C_1|^2 + \frac{3}{8} |B_0|^2 + \frac{1}{8} |C_0|^2 + \\ &\quad + \frac{1}{4\sqrt{3}} |B_1| |C_1| \cos \delta + \frac{\sqrt{3}}{4} |B_0| |C_0| \cos \delta, \\ \sigma(K^- + pp \rightarrow \Sigma^+ + N) &\approx \frac{1}{9} |B_1|^2 + \frac{1}{3} |C_1|^2 - \frac{2}{3\sqrt{3}} |B_1| |C_1| \cos \delta, \\ \sigma(K^- + pp \rightarrow \Sigma^0 + p) &\approx \frac{1}{2} \sigma(K^- + pp \rightarrow \Sigma^+ + N), \\ \sigma(K^- + pp \rightarrow \Lambda^0 + p) &\approx \frac{1}{2} |B_1|^2 + \frac{1}{6} |C_1|^2 + \frac{1}{\sqrt{3}} |B_1| |C_1| \cos \delta, \\ \sigma(K^- + d \rightarrow \Sigma^- + p) &\approx \frac{1}{6} |B_0|^2 + \frac{1}{2} |C_0|^2 - \frac{1}{\sqrt{3}} |B_0| |C_0| \cos \delta, \\ \sigma(K^- + d \rightarrow \Sigma^0 + N) &\approx \frac{1}{2} \sigma(K^- + d \rightarrow \Sigma^- + p), \\ \sigma(K^- + d \rightarrow \Lambda^0 + N) &\approx \frac{3}{4} |B_0|^2 + \frac{1}{4} |C_0|^2 + \frac{\sqrt{3}}{2} |B_0| |C_0| \cos \delta, \end{aligned} \right.$$

where $\delta = |\alpha_1 - \alpha_0|$ the difference of the triplet and singlet nucleon-nucleon phase shift. A little examination of (9) establishes the following equalities between K^+ -multinucleon and deuteron reaction rates,

$$(10) \quad \sigma(K^- + pN \rightarrow \Sigma^- + p) = \frac{1}{4} \sigma(K^- + pp \rightarrow \Sigma^+ + N) + \frac{1}{2} \sigma(K^- + d \rightarrow \Sigma^- + p),$$

$$(11) \quad \sigma(K^- + pN \rightarrow \Lambda^0 + N) = \frac{1}{4} \sigma(K^- + pp \rightarrow \Lambda^0 + p) + \frac{1}{2} \sigma(K^- + d \rightarrow \Lambda^0 + N).$$

The method we have adopted here in obtaining the various cross-sections was used by AMATI and VITALE for the ordinary $K^- + p$ reactions. They have (predicted by global symmetry) proposed a possible inequality among the various cross-sections. This has been tested ⁽⁹⁾ subsequently with the $K^- + p$ reactions data with the conclusion that global symmetry is false. We have not predicted here any such inequality among the various $K^- + (2N)$ reaction cross-sections. On the other hand we analyse the data in the following way:

From the $K^- + d$ absorption data ⁽¹⁰⁾ at low energy we have the branching ratio for charged to neutral hyperon (Λ^0, Σ^0) production as

$$(12) \quad \sigma(\Lambda^0) + \sigma(\Sigma^0) : \sigma(\Sigma^-) = 7:8.$$

The separation of Σ^0 and Λ^0 seems very difficult at the moment. Although the multinucleon reaction rates are not very clear, the experiment seems to indicate ⁽¹¹⁾ that the following branching ratios.

$$\frac{\sigma(K^- + pN \rightarrow \Sigma^- + p)}{\sigma(K^- + pp \rightarrow \Sigma^+ + N)} \quad \text{and also} \quad \frac{\sigma(K^- + pN \rightarrow \Lambda^0 + N)}{\sigma(K^- + pp \rightarrow \Lambda^0 p)},$$

are the same and ≈ 1.3 . A fit to these experimental situations with the help of the cross-sections already calculated in (9), (10) and (11) requires that $\cos \delta < 0$ indicating that $90^\circ < \delta < 270^\circ$.

On the otherhand the nucleon-nucleon scattering experiment at low energy indicates that (*s*-wave phase shift is predominantly large) singlet triplet phase shift difference ⁽¹²⁾ namely $\delta (= \alpha_1 - \alpha_0) \approx 23^\circ$ only and at still higher energy the higher angular momentum singlet-triplet phase shift difference is very

⁽⁹⁾ D. AMATI and B. VITALE: *Nuovo Cimento*, **9**, 895 (1958); R. H. DALITZ: *Ann. International Conference on High Energy Physics at CERN* (1958), p. 187.

⁽¹⁰⁾ Report by ALVAREZ in *High Energy Physics Conference at Kiev* (1959).

⁽¹¹⁾ G. ALEXANDER, Y. EISENBERG and D. KESSLER: *Nuovo Cimento*, **15**, 484 (1960).

⁽¹²⁾ *High Energy Nuclear Physics Conference at Rochester* (1957), p. III-12.

small. Thus we conclude that the particular restricted symmetry (included in the global symmetry hypothesis) is obviously incompatible with the present day experimental data on K^- absorption in nuclei. It must be pointed out that this result is not based on the perturbative approach at all.

Rigorously speaking when one compares the phase-shift difference $\delta(=\alpha_1 - \alpha_0)$ as predicted by our theoretical analysis, with the experimental observation one must definitely know in which angular momentum states the final state particles are produced. In view of the several particles present one would encounter the occurrence of various angular momentum states. Further the problem of assigning definite final angular momentum states is very much complicated because of the lack of definite knowledge about the relative hyperon, nucleon and K-meson parities and also one must bear in mind that in all reactions the final states being hyperons and nucleons, Pauli exclusion principle is not operative which would otherwise reduce the number of possible angular momentum states. However, assuming parity and angular momentum conservation and with a definite choice of either of the possibilities of $K^+\mathcal{N}^+\Sigma^+$, $K^+\mathcal{N}^+\Lambda^+$, $K^+\mathcal{N}^+\Sigma^+$ and $K^-\mathcal{N}^+\pi^+$ relative parity (where the notation is the same as that of GELL-MANN⁽¹³⁾) we have a simple kinematical analysis of the various $K^- + d$ reactions. It is simpler because in these one knows in advance that the deuteron has $J=1$ with triplet spin states. The situation becomes complicated for $K^- + 2\mathcal{N}$ reactions when $2\mathcal{N}$ system may have all possible values of the angular momentum L . Since the reactions are observed at a very low energy one can assume that the K-meson is absorbed in an S -state. Confining our attention only to $L=0$ and $l=0$, where l is the angular momentum of K^- -meson with respect to the centre of mass of the 2-nucleon system, we notice, that if the relative parity of K-meson-hyperon be odd, then the final hyperon-nucleon states would be produced in 3P_1 and 1P_1 states for deuteron reactions, for $(p\mathcal{N})$ reactions they would be in $^3P_{1,0}$ and 1P_1 states and in 3P_0 states for (pp) reactions. If the K-H relative parity is even then the final states are evidently in 1S_0 or 3S_1 states. Hence in comparing our prediction we should take the P -wave $\mathcal{N}\text{-}\mathcal{N}$ phase-shift of the relative K-H parity odd or the S -wave, $\mathcal{N}\text{-}\mathcal{N}$ phase shift for even relative K-H parity.

It may be pointed out that 3P_1 and 1P_1 ($\mathcal{N}\text{-}\mathcal{N}$) phase shifts are both quite equally small and of the same sign while 3P_0 phase shift is of opposite sign but is very small. Then it would be sufficiently justified if we consider one of these phases. If we take into account all of them this will only increase the number of parameters which would not be possible to fit with the relatively scanty data available now of the K^- absorption processes.

(13) M. GELL-MANN: *Phys. Rev.*, **106**, 1296 (1957).

* * *

Our thanks are due to Mr. B. M. UDGAONKAR and Drs. L. K. PANDIT and V. GUPTA for discussions.

RIASSUNTO (*)

Il principio di indipendenza dalla carica associato alla unitarietà e reversibilità nel tempo della matrice di scattering permette di esprimere l'ampiezza di produzione delle reazioni di cattura K^- in termini delle fasi di scattering delle interazioni finali di stato. Applicando approssimazioni del tipo dell'impulso, si ottengono alcune relazioni fra le sezioni trasversali di assorbimento $K^- + d$ e $K^- + 2N$. I dati sperimentali sono stati analizzati sulla base della simmetria globale delle interazioni forti. Si trova che entro i limiti della nostra approssimazione questa simmetria è incompatibile con le osservazioni sperimentali.

(*) Traduzione a cura della Redazione.

On the Decay Rate of the Charged Pion.

J. BEENSTEIN (*)

Faculté des Sciences - Orsay

S. FUBINI

Istituto di Fisica dell'Università - Padova
CERN - Geneva

M. GELL-MANN (**)

*Collège de France and Ecole Normale Supérieure - Paris (***)*

W. THIRRING

Institute for Theoretical Physics, University of Vienna - Vienna
CERN - Geneva

(ricevuto il 7 Giugno 1960)

Summary. – The Goldberger-Treiman relation between the pion decay amplitude and the axial vector coupling constant in β -decay is shown to be plausible if the divergence of the axial vector current is a highly non-singular operator (*i.e.*, one that emphasizes low frequencies). Such a situation is probably realized, for example, in a renormalizable theory for which this operator is proportional to the conventional pion field. Some other possible consequences of the low-frequency character of the operator are mentioned.

1. – Introduction.

GOLDBERGER and TREIMAN ⁽¹⁾ have stated a formula for the rate of charged pion decay that is in excellent agreement with experiment. Their derivation of the formula was based, however, on a number of assumptions, some of

(*) National Science Foundation Post-Doctoral Fellow.

(**) National Science Foundation Senior Post-Doctoral Fellow.

(***) Permanent address: California Institute of Technology, Pasadena, Cal., U.S.A.

(1) M. L. GOLDBERGER and S. B. TREIMAN: *Phys. Rev.*, **110**, 1478 (1958). See also M. L. GOLDBERGER: *Rev. Mod. Phys.*, **31**, 797 (1959).

which are not easy to justify. Various authors ⁽²⁻⁴⁾ have therefore tried to find other chains of argument that would lead, at least approximately, to the same formula. Some of these attempts have involved abandoning the conventional form of the strong pion-nucleon interaction or the conventional form of the weak axial vector current or both.

Recently ⁽⁵⁾ the possibility has been considered that the strong interactions and the weak current are such that the divergence of the axial vector current P_α (for baryons and mesons with $\Delta S = 0$) is proportional to a component of the pion field. Under that condition the G-T formula can be made particularly plausible. A number of models of strong and weak interactions possessing the property in question were presented, but none of them was free of difficulties.

It is interesting, therefore, to see if the kind of argument that was used in A to obtain the G-T formula can be made more general. It is evident at once that there is something artificial about the requirement that $\partial_\alpha P_\alpha$ be proportional to π , since it is somewhat arbitrary which pseudoscalar field with $S = 0$ and $I = 1$ is called the pion field. With this in mind, let us try to isolate, in a way that does not involve a choice of which field is to be called π , the assumptions in A that are relevant to the deduction of the G-T formula.

2. - The essence of the G-T relation.

Whatever arbitrariness there may be in the definition of the renormalized pion field operator π_r , its matrix elements between one-pion states and the vacuum are uniquely defined. The corresponding matrix element of the divergence of the axial vector current can always be written in the form

$$(1) \quad \langle 0 | \partial_\alpha P_\alpha | \pi^- \rangle = i a_1 2^{-\frac{1}{2}} \langle 0 | \pi_r^- | \pi^- \rangle,$$

where the constant a_1 corresponds to what was called $a\sqrt{Z_3}$ in A. The pion decay rate is then

$$(2) \quad \Gamma_\pi = \frac{G^2}{16\pi m_\pi} \frac{m_\mu^2}{m_\pi^2} \left(1 - \frac{m_\mu^2}{m_\pi^2} \right)^2 a_1^2,$$

where we have chosen the phases so that a_1 is real.

⁽²⁾ R. F. STREATER and J. C. TAYLOR: *Nuclear Physics*, **7**, 276 (1958).

⁽³⁾ R. E. NORTON and W. K. R. WATSON: *Phys. Rev.*, **110**, 996 (1958).

⁽⁴⁾ R. F. SAWYER: *Phys. Rev.*, **116**, 231 (1959).

⁽⁵⁾ M. GELL-MANN and M. LÉVY: *Nuovo Cimento*, **16**, 705 (1960), to be referred to as A; and J. BERNSTEIN, M. GELL-MANN and L. MICHEL: *Nuovo Cimento*, **16**, 560 (1960), to be referred to as B. We shall employ the notation of these articles and we shall quote equations from them as (A-1), (B-2), etc.

The operator λ^- defined by the equation

$$(3) \quad \lambda^- \equiv -i\sqrt{2}a_1^{-1}\partial_\alpha P_\alpha$$

evidently behaves like the renormalized pion field π_r^- between one-pion states and the vacuum and may always be thought of as a kind of candidate for the title of pion field. It was suggested in A that the condition $\lambda^- = \pi_r^-$ would make the G-T relation plausible. Whether or not this is so, the argument is that if certain properties that might reasonably be attributed to the operator π_r^- are possessed by the « candidate » λ^- , then the G-T relation could be understood in a simple way. We must try to specify, as nearly as possible, what are the desirable properties of λ^- .

We are especially interested in the matrix-element of λ^- between one-nucleon states. Putting, as in (B-18),

$$(4) \quad \langle p | P_\alpha | n \rangle = -\frac{G_A}{G} \alpha(k^2) \bar{u}_f \tau_+ \gamma_\alpha \gamma_5 u_i + i k_\alpha \beta(k^2) \bar{u}_f \tau_+ \gamma_5 u_i,$$

and defining K_λ by the relation

$$(5) \quad \langle p | \lambda^- | n \rangle = i\sqrt{2} \bar{u}_f \tau_+ \gamma_5 u_i K_\lambda(k^2),$$

we have

$$(6) \quad 2m \left(-\frac{G_A}{G} \right) \alpha(k^2) + k^2 \beta(k^2) = a_1 K_\lambda(k^2).$$

Since $\alpha(0)$ is unity by definition, the formula relating the axial vector coupling constant to the pion decay amplitude is just

$$(7) \quad -\frac{G_A}{G} = \frac{a_1}{2m} K_\lambda(0).$$

Now the function K_λ is the same one that occurs in the creation of a nucleon-antinucleon pair from the vacuum by the operator λ^- , *i.e.*, in the matrix element $\langle p, \bar{n} | \lambda^- | 0 \rangle$. As remarked in A, the function always has a pole at $k^2 = -m_\pi^2$ with residue $-g_1$, where g_1 is the renormalized pion-nucleon coupling constant, because λ^- behaves as the renormalized pion field for the creation of real pions from the vacuum. This situation is perfectly general and was pointed out long ago ⁽⁶⁾ by GOLDBERGER and TREIMAN and others

⁽⁶⁾ M. L. GOLDBERGER and S. B. TREIMAN: *Phys. Rev.*, **111**, 355 (1958); L. WOLFENSTEIN: *Nuovo Cimento*, **8**, 882 (1958); J. L. LOPES: *Phys. Rev.*, **109**, 509 (1958).

in their work on the induced pseudoscalar term $\beta(k^2)$. The pole in question, coming from the intermediate one-pion state in the sense of dispersion theory, occurs only in the β term of eq. (6). We have, then, in any theory,

$$(8) \quad K_\lambda(k^2) \approx \frac{-g_1}{k^2 + m_\pi^2} \quad \text{for } k^2 \approx -m_\pi^2,$$

or, what is the same thing,

$$(9) \quad K_\lambda(k^2) = \frac{-g_1}{k^2 + m_\pi^2} \varphi_\lambda(k^2) \quad \text{where } \varphi_\lambda(-m_\pi^2) = 1.$$

In this language, the G-T relation is simply the statement that $\varphi_\lambda(k^2)$ remains close to unity between $-m_\pi^2$ and 0 so that $\varphi_\lambda(0) \approx 1$. If φ_λ is slowly varying in this way, then we have

$$(10) \quad K_\lambda(0) \approx \frac{-g_1}{m_\pi^2}$$

and thus

$$(11) \quad -G_A/G \approx \frac{-a_1 g_1}{2mm_\pi^2},$$

which is the G-T relation.

In order to explain the pion lifetime, all that must be established in any theory of the strong and weak couplings is that the one-pion pole in K_λ continues to dominate at $k^2 = 0$.

Similarly, in order to derive the prediction of GOLDBERGER and TREIMAN for the induced pseudoscalar term in μ^- capture by protons, all that must be established is that the one-pion pole in $\beta(k^2)$

$$(12) \quad \beta(k^2) \approx \frac{a_1 g_1}{m_\pi^2} \frac{1}{k^2 + m_\pi^2} \quad \text{for } k^2 \approx -m_\pi^2,$$

continues to dominate at $k^2 = +m_\mu^2$:

Besides the pole at $k^2 = -m_\pi^2$, the functions β and K_λ have branch lines running from $-9m_\pi^2$ to $-\infty$, which represent the contributions of the intermediate states other than the one-pion state, in the sense of dispersion theory. Either function, if it vanishes at infinity, may be written simply as the sum of an integral over the branch line and a term representing the pole. In other words, we have a dispersion relation without subtractions. For instance, if

$K_\lambda \rightarrow 0$ as $|k^2| \rightarrow \infty$, then we have

$$(13) \quad K_\lambda(k^2) = \frac{-g_1}{k^2 + m_\pi^2} + \int_{9m_\pi^2}^{\infty} dM^2 \frac{\sigma(M^2)}{k^2 + M^2}.$$

If, however, K_λ fails to approach zero at infinity, then we must make a subtraction in the dispersion relation (13) and the value of K_λ at some particular value of k^2 , such as $k^2 = 0$, become an arbitrary constant.

The weight function σ (which is not necessarily positive) is the sum of products of two factors, one representing the amplitude for the creation of a state of mass M from the vacuum by the operator λ^- and the other representing the transition amplitude from such a state to a final state of proton and antineutron.

One way to understand the success of the G-T relation, then, is to suppose that λ is an operator which, acting on the vacuum, emphasizes low frequencies, in such a way that:

a) The matrix elements for creation of states of high mass tend to zero as the mass goes to infinity; in particular K_λ , which represents $\langle p, \bar{n} | \lambda^- | 0 \rangle$, tends to zero⁽⁷⁾, giving the unsubtracted dispersion relation (13). (We note that if $K_\lambda \rightarrow 0$ and if α is not to increase like k^2 at infinity, then $\beta \rightarrow 0$ and obeys an unsubtracted dispersion relation).

b) The matrix elements for creation of higher states than the pion are not large, so that, for normal values of the transition amplitudes mentioned above, the one pion term $-g_1/m_\pi^2$ dominates the integral

$$\int_{9m_\pi^2}^{\infty} \sigma(M^2) \frac{dM^2}{M^2}.$$

This point of view is not without experimental implications, although the experiments involved are difficult to perform at the present time. For example, we should expect the one-pion pole to predominate near $k^2 = 0$ in the case of the matrix elements $\langle \Sigma^+, \bar{A} | \lambda^- | 0 \rangle$ and $\langle \Lambda, \bar{\Sigma}^- | \lambda^- | 0 \rangle$, giving an approximate relation like the G-T formula linking the renormalized pion coupling constant for $\Lambda \leftrightarrow \Sigma$ transitions to the effective axial vector coupling constants for $\Sigma^+ \rightarrow \Lambda + e^+ + \nu$ and $\Sigma^- \rightarrow \Lambda + e^- + \bar{\nu}$ (or, more accurately, the average of these two.)

(7) This crucial point has recently been made independently by Y. NAMBU: *Phys. Rev. Lett.*, **4**, 380 (1960).

If we apply the same idea to the matrix element $\langle \mathcal{N}, \overline{\mathcal{N}}, \pi | \lambda^- | 0 \rangle$, we obtain an approximate prediction for small k^2 of the value of the axial vector amplitude for processes like $\bar{\nu}' + \mathcal{N} \rightarrow \mu^+ + \mathcal{N} + \pi$, in terms of the pion-nucleon scattering amplitude.

In summary, then, what we suppose to be at the root of the G-T relation is that λ^- is a highly non-singular operator that emphasizes low frequencies. In the limit of high frequencies, then, the axial vector current is conserved.

The formal equality $\lambda^- = \pi_r^-$ (where π_r^- is a conventionally defined pion field) is useful in that, in a renormalizable theory like the σ model of article A it makes plausible the non-singular character of λ^- .

3. - The propagator.

For a conventional pion field π_r^- , it is customary to consider the renormalized pion propagator $0 | P(\pi_r^-(x), \pi_r^+(y)) | 0 \rangle$ with Fourier transform ⁽⁸⁾

$$(14) \quad \frac{d_\pi(k^2)}{k^2 + m_\pi^2 - i\varepsilon} = \frac{1}{k^2 + m_\pi^2 - i\varepsilon} + \int_{9m_\pi^2}^{\infty} \frac{\varrho_\pi(M^2) dM^2}{k^2 + M^2 - i\varepsilon}$$

and to suppose that the integral converges, i.e., that the pion propagator in momentum space is finite.

For the operator λ_r^- , we may also construct a propagator $\langle 0 | P(\lambda_r^-(x), \lambda_r^+(y)) | 0 \rangle$ and write its Fourier transform formally as

$$(15) \quad \frac{d_\lambda(k^2)}{k^2 + m_\pi^2 - i\varepsilon} = \frac{1}{k^2 + m_\pi^2 - i\varepsilon} + \int_{9m_\pi^2}^{\infty} \frac{\varrho_\lambda(M^2) dM^2}{k^2 + M^2 - i\varepsilon}.$$

If λ is a sufficiently non-singular operator so that it gives a finite propagator in momentum space, then it is true *a fortiori* that $K_\lambda \rightarrow 0$ at infinity. We may see this by writing $\varrho_\lambda(M^2) \geq \varrho_\lambda^{\text{pair}}$, where $\varrho_\lambda^{\text{pair}}$ is the contribution to the weight function ϱ_λ from states containing one nucleon and one antinucleon and is given by

$$(16) \quad \varrho_\lambda^{\text{pair}} = (8\pi^2)^{-1} [M^2(M^2 - 4m^2)]^{\frac{1}{2}} (M^2 - 4m^2) |K_\lambda(-M^2)|^2.$$

⁽⁸⁾ G. KÄLLÉN: *Helv. Phys. Acta*, **25**, 417 (1952). See also M. GELL-MANN and F. E. LOW: *Phys. Rev.*, **95**, 1300 (1954); H. LEHMANN: *Nuovo Cimento*, **11**, 342 (1954).

Then we have

$$(17) \quad m_{\pi}^{-2} d_{\lambda}(0) \geq m_{\pi}^{-2} + (8\pi^2)^{-1} \int_{4m^2}^{\infty} dM^2 |K_{\lambda}(-M^2)|^2 \left(1 - \frac{4m^2}{M^2}\right)^{\frac{1}{2}},$$

which, of course, forces K_{λ} to go to zero at infinity. Even if $d_{\lambda}(0) - 1 = m_{\pi}^2 \int (\varrho_{\lambda} dM^2)/M^2$ diverges, but not so badly as to make the next moment $\int (\varrho_{\lambda}(M^2)/M^4) dM^2$ diverge as well, that is sufficient to make $K_{\lambda} \rightarrow 0$.

It is interesting that we can formally calculate $d_{\lambda}(0)$, using the definition (3) of λ^- as proportional to the divergence of the axial vector current. We find

$$(18) \quad \langle 0 | P(\lambda^-(x), \lambda^+(y)) | 0 \rangle = \\ = -i\sqrt{2} a_1^{-1} \left\{ \frac{\partial}{\partial x_{\alpha}} \langle 0 | P(P_{\alpha}(x), \lambda^+(y)) | 0 \rangle + i\delta(x_0 - y_0) \langle 0 | [P_4(x), \lambda^+(y)] | 0 \rangle \right\}.$$

We may put

$$(19) \quad [P_4(\mathbf{x}, x_0), \lambda^+(\mathbf{y}, x_0)] \equiv \delta(\mathbf{x} - \mathbf{y})(-i2^{-\frac{1}{2}})X(\mathbf{x}, x_0).$$

Going over to the Fourier transform $(k^2 + m_{\pi}^2)^{-1} d_{\lambda}(k^2)$, we remark that the divergence term in (18) contributes nothing at $k=0$, so that we have

$$(20) \quad m_{\pi}^{-2} d_{\lambda}(0) = \langle 0 | X | 0 \rangle a_1^{-1}.$$

In the σ model considered in A, X is simply $2aa_1^{-1}\sigma' = 2\sigma'Z_3^{-\frac{1}{2}}$, so that we have

$$m_{\pi}^{-2} d_{\lambda}(0) = m_{\pi}^{-2} d_{\pi}(0) = 2a_1^{-1} \langle 0 | \sigma' Z_3^{-\frac{1}{2}} | 0 \rangle.$$

According to the σ model, the quantities $d_{\pi}(0)$, a_1 , and $\langle 0 | \sigma' Z_3^{-\frac{1}{2}} | 0 \rangle$ are all finite, at least in the series expansion of perturbation theory.

In the gradient coupling model, X is just the number $\mu_0^2 f_0^{-2} a_1^{-1}$. Since in that model a_1 is $-Z_3^{\frac{1}{2}} \mu_0^2 f_0^{-1}$, eq. (20) becomes

$$m_{\pi}^{-2} d_{\lambda}(0) = m_{\pi}^{-2} d_{\pi}(0) = \mu_0^{-2} Z_3^{-1},$$

which we know to be true for the gradient coupling model. (See eq. (A-35)).

In the usual pseudoscalar pion theory with pseudoscalar coupling to nucleons and with P_{α} taken to be simply $\bar{p}\gamma_{\alpha}\gamma_5 n$, we obtain formally

$$X = -4a_1^{-1}(m_0 \bar{\mathcal{N}}\mathcal{N} - ig_0 \bar{\mathcal{N}}\gamma_5 \boldsymbol{\tau} \cdot \boldsymbol{\pi} \mathcal{N}) = 4a_1^{-1} \bar{\mathcal{N}}\gamma_{\alpha} \partial_{\alpha} \mathcal{N},$$

and it looks as if $\langle X \rangle$ and $d_{\lambda}(0)$ are highly divergent. Of course that does not exclude the possibility of K_{λ} going to zero any way.

4. — Correspondance with the arguments of G-T.

In a theory in which λ^- is not the conventional π_r^- field and in which we cannot establish that λ^- gives a sensible pion propagator, we may try to get some information about K_λ by looking at a mixed propagator, which is neither

$$\langle 0 | P(\pi_r^-(x), \pi_r^+(y)) | 0 \rangle$$

nor

$$\langle 0 | P(\lambda^-(x), \lambda^+(y)) | 0 \rangle,$$

but

$$\frac{1}{2} \left(\langle 0 | P(\lambda^-(x), \pi_r^+(y)) | 0 \rangle + \langle 0 | P(\pi_r^-(x), \lambda^+(y)) | 0 \rangle \right).$$

The Fourier transform of the first propagator gives, of course

$$(21a) \quad \frac{d_\pi(k^2)}{k^2 + m_\pi^2 - i\varepsilon} = \frac{1}{k^2 + m_\pi^2 - i\varepsilon} + \int_{4m^2}^{\infty} \frac{\sqrt{(M^2 - 4m^2)M^2} dM^2 |K_\pi(-M^2)|^2}{8\pi^2 (k^2 + M^2 - i\varepsilon)} + \dots,$$

where the remaining terms, like the one-pion and one-pair terms, have positive weight functions. Likewise the second propagator gives, in Fourier transform

$$(21b) \quad \frac{d_\lambda(k^2)}{k^2 + m_\pi^2 - i\varepsilon} = \frac{1}{k^2 + m_\pi^2 - i\varepsilon} + \int_{4m^2}^{\infty} \frac{\sqrt{(M^2 - 4m^2)M^2} dM^2 |K_\lambda(-M^2)|^2}{8\pi^2 (k^2 + M^2 - i\varepsilon)} + \dots,$$

with positive weight functions everywhere. The mixed propagator gives

$$(21c) \quad \frac{d_{\text{mixed}}(k^2)}{k^2 + m_\pi^2 - i\varepsilon} = \frac{1}{k^2 + m_\pi^2 - i\varepsilon} + \int_{4m^2}^{\infty} \frac{\sqrt{M^2(M^2 - 4m^2)} dM^2 \operatorname{Re} K_\lambda(-M^2) K_\pi^*(-M^2)}{8\pi^2 (k^2 + M^2 - i\varepsilon)},$$

where the weight functions need not be positive.

Now just as we showed that $m_\pi^{-2} d_\lambda(0) = a_1^{-1} \langle 0 | X | 0 \rangle$, we can define

$$(22) \quad [P_4(\mathbf{x}, x_0), \pi_r^+(\mathbf{y}, x_0)] = \delta(\mathbf{x} - \mathbf{y}) \left(-\frac{i}{\sqrt{2}} \right) Y,$$

and obtain

$$(23) \quad m_\pi^{-2} d_{\text{mixed}}(0) = \langle 0 | Y | 0 \rangle a_1^{-1}.$$

In the simple pseudoscalar theory with $P_\alpha = \bar{P}\gamma_\alpha\gamma_5 n$, the operator Y vanishes and so we obtain

$$0 = \frac{1}{m_\pi^2} + \int_{4m^2}^{\infty} \frac{\sqrt{M^2(M^2 - 4m^2)}}{8\pi^2} \frac{dM^2}{M^2} \operatorname{Re} K_\lambda(-M^2) K_\pi^*(-M^2) + \dots,$$

which is the starting point of the method used by GOLDBERGER and TREIMAN to derive their relation. Using eq. (9) we may rewrite the equation in the form

$$(25) \quad 0 = \frac{1}{m_\pi^2} + g_1^2 \int_{4m^2}^{\infty} \frac{\sqrt{M^2(M^2 - 4m^2)}}{8\pi^2} \frac{dM^2}{M^2} \frac{\operatorname{Re} \varphi(-M^2) q_\pi^*(-M^2)}{(M^2 - m_\pi^2)^2} + \dots$$

Unfortunately, the absence of any requirement of positiveness means that we cannot use inequalities. GOLDBERGER and TREIMAN therefore are obliged to throw away the remaining terms, such as contributions from 3π states, and an unknown error is thereby introduced.

Even if we do throw away the other terms, we do not have clear sailing since we do not know the functional forms of q_λ or φ_π : GOLDBERGER and TREIMAN get around these difficulties by two more assumptions:

1) They estimate φ_π by a dispersion theory calculation using one-pion and one-pair states only.

2) They assume a simple trial form for φ_λ , namely $1 + ((k^2 m_\pi^2) y / m_\pi^2)$ where y is a number. (Note that K_λ then approaches a constant at infinity, rather than zero, which would be our conjecture. This is not a very serious difference between the points of view, because the trial form need hold only up to a few GeV in the work of G-T, not necessarily all the way to infinity.)

Finally they use eq. (32) to calculate y and find that it is small; it follows that $\varphi_\lambda(0)$ is close to 1, which is of course, their important result.

5. - Conclusions.

The success of the G-T formula for the rate of pion decay indicates that the function K_λ is dominated near $k^2 = 0$ by its one pion pole. Probably the operator $\hat{\lambda}^-$ is sufficiently non-singular so that K_λ vanishes as $k^2 \rightarrow \infty$.

If so, it is possible that λ is even more non-singular and that λ , treated as the renormalized pion field, gives a pion propagator with a convergent spectral representation. It is a description of such a situation that was attempted in

A and B, particularly in the σ model. If the situation obtains, then the vacuum expectation value of the commutator of P_4 and λ must be finite.

That λ be a suitable pion field is, however, only a sufficient condition for the vanishing of K_λ at ∞ ; it is not necessary, however, and we may if we like, concentrate on the implications of the dominance of the one-pion pole as the explanation of the G-T formula.

First, there is no uncertainty in the relative sign of the axial vector term $-G_A \alpha(k^2)$ and the induced pseudoscalar term $G\beta(k^2)$. (In the problem of muon capture, the possibility of varying the relative sign has been discussed⁽⁹⁾; crude comparisons of theory and experiment favor the sign given above, which is also the original sign of GOLDBERGER and TREIMAN.)

Second, the matrix element of λ between Σ and Λ may also be dominated by the one-pion pole at small k^2 . Thus we can relate the strength of the axial vector interaction in the decay $\Sigma \rightarrow \Lambda + \text{leptons}$ to the value of the renormalized coupling constant for the pion to Σ and Λ .

Third, the matrix element of λ between a nucleon state and a state with one nucleon and one pion may be given mainly by the one-pion pole. That would permit the calculation of the axial vector amplitude for $\nu + p \rightarrow \pi^0 + e^+ + n$, etc., at small momentum transfers.

A discussion of these points and of the role of the one-pion pole in nuclear β -transitions will be given elsewhere⁽¹⁰⁾.

⁽⁹⁾ See especially H. PRIMAKOFF: *Rev. Mod. Phys.*, **31**, 802 (1959).

⁽¹⁰⁾ Note added in proof: Results analogous to ours have been obtained independently by Chou Kuang-Chao - Dubna Report D 514 (1960).

RIASSUNTO

Si dimostra la plausibilità della relazione di Goldberger e Treiman fra vita media del pione e costante d'accoppiamento assiale nel caso in cui la divergenza della corrente pseudovettoriale è altamente non-singolare (cioè che contiene in prevalenza basse frequenze). Tale situazione ha probabilmente luogo per esempio in una teoria rinormalizzabile in cui quest'operatore è proporzionale al campo mesonico. Si discutono altre possibili conseguenze del carattere non singolare dell'operatore.

Form-Factor of the Neutron from Deuteron Electro-Disintegration.

S. K. BOSE

*Department of Physics and Astronomy - University of Rochester
Rochester, N.Y.*

(ricevuto il 9 Giugno 1960)

Summary. — An extrapolation method is suggested to determine the magnetic form-factor of the neutron from deuteron electro-disintegration.

The inelastic electron-deuteron scattering process $e+d \rightarrow e+n+p$ has recently received both experimental and theoretical attention ⁽¹⁾. The principal interest in these studies lies in the possibility of determining the form-factor of the neutron from an analysis of the observed inelastic scattering cross-section.

At the same time, following the suggestion by CHEW ⁽²⁾ to find the π -n coupling constant from n-n scattering, attempts have been made to determine physically interesting quantities based on the recently discussed analytic properties of transition amplitudes. These studies embrace the method of determining the π - π coupling constant from inelastic π -n scattering ⁽³⁾, the determination of the π -n coupling constant from pion photo-production angular distribution ⁽⁴⁾ and a method to determine the form-factor of the pion from pion electro-production experiments ⁽⁵⁾.

⁽¹⁾ The latest calculation on the subject is due to L. DURAND III: *Phys. Rev.*, **115**, 1020 (1959) which contains reference to earlier works.

⁽²⁾ G. F. CHEW: *Phys. Rev.*, **112**, 1380 (1958).

⁽³⁾ C. GOEBEL: *Phys. Rev. Lett.*, **1**, 337 (1958).

⁽⁴⁾ J. G. TAYLOR, M. J. MORAVCSIK and J. L. URETSKY: *Phys. Rev.*, **113**, 689 (1959).

⁽⁵⁾ W. R. FRAZER: *Phys. Rev.*, **115**, 1761 (1959).

The purpose of the present communication is to point out a method for determining the neutron form-factor from the inelastic electron-deuteron scattering, exploiting the same principle as that of the last mentioned authors.

The method is based upon a procedure of extrapolation of the deuteron electro-disintegration cross-section. The basic principle involved is the possibility of analytic continuation of the electro-disintegration cross-section as a function of the square of the four-momentum transfer between the deuteron and one of the final nucleons, Δ^2 . We denote the initial four-momentum of the deuteron by p , that of the final proton and neutron by p' and q respectively. Δ^2 is then $-(p-p')^2$. The inelastic scattering amplitude will have a pole at $\Delta^2 = -m^2$ when m is the nucleon mass. The residue of the pole is just the electromagnetic form-factor of the neutron multiplied by a known coefficient. The pole occurs, of course, in the unphysical region and hence, an extrapolation of the observed cross-section will be needed to reach it.

The analytic properties of the electro-disintegration amplitude, on which the proposed extrapolation procedure is based, have not been proved so far. They have, however, been often conjectured, for instance, in the two-dimensional spectral representation proposed by MANDELSTAM ⁽⁶⁾.

Using the recipe advocated in the paper referred to, we can derive the singularities of the electro-disintegration amplitude in the following way. Consider the spectrum of intermediate states that can be reached if p and p' were incoming and q and k outgoing. k is the four-momentum transfer between electrons, $k = s - s'$, where s and s' denote the initial and final electron four-momenta. The lowest state, i.e. the state with the least mass, that can be reached is a one nucleon state. This gives a pole at $\Delta^2 = -m^2$. The next is the pion-nucleon state which gives a branch point at $\Delta^2 = -(m+\mu)^2$ μ being the pion mass.

The other set of singularities coming from the «crossed» spectrum is obtained by considering p and q incoming and p' and k outgoing. This gives a pole at $(p-q)^2 = m^2$ and a branch point at $(p-q)^2 = (m+\mu)^2$. The singularities can be re-expressed in terms of Δ^2 using the relation $(p-q)^2 = \Delta^2 - E^2 + 2m^2 + M^2 - \lambda$ obtained from the conservation relation $p + s = p' + q + s'$; one obtains a pole at $\Delta^2 = \lambda - m^2 - M^2 + E^2$ and a branch point at $\Delta^2 = \mu^2 + 2m\mu - m^2 - M^2 + E^2 + \lambda$ where $E^2 = (p' + q)^2$ i.e. the total energy of the nucleons in their barycentric system. M is the deuteron mass and $\lambda = -k^2$.

The discussion on the singularities of the electro-disintegration amplitude given above is, however, incomplete in one important respect. This is because of the appearance of additional singularities referring to the structure of the deuteron as a weakly-bound composite system. These anomalous threshold

⁽⁶⁾ S. MANDELSTAM: *Phys. Rev.*, **112**, 1344 (1959).

singularities give branch points at

$$\Delta^2 = -m^2 - \frac{M^2}{2m^2} N^2 \mu^2 - \frac{M\mu N}{m} \sqrt{4m^2 - M^2} \sqrt{1 - N^2 \mu^2 / 4m^2}$$

for the uncrossed spectrum and branch points at

$$\Delta^2 = E^2 + \lambda - M^2 - m^2 + \frac{M^2 \mu^2 N^2}{2m^2} + \frac{MN\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - \frac{N^2 \mu^2}{4m^2}},$$

for the crossed spectrum. These singularities are discussed in a separate Appendix.

The spectrum of singularities may also be exhibited in the $\cos \theta$ plane, where θ is the angle between \mathbf{k} and \mathbf{q} in the $\mathbf{p}' + \mathbf{q} = 0$ frame. The pole occurs at

$$\cos \theta = \frac{\lambda + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}$$

and the branch point at

$$\cos \theta = \frac{\lambda + 2m\mu + \mu^2 + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}.$$

The «crossed» pole and branch point occur at

$$\cos \theta = \frac{M^2 - E^2 + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}$$

and at

$$\cos \theta = \frac{M^2 - E^2 - 2m\mu - \mu^2 + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}.$$

The first anomalous threshold branch point for the «uncrossed» and crossed spectra occur at

$$\cos \theta = \frac{\lambda + 2\mu^2 + 4\mu\gamma}{2|\mathbf{k}||\mathbf{q}|}$$

and at

$$\cos \theta = \frac{M^2 - 2\mu^2 - 4\mu\gamma - E^2 + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}$$

respectively, where $\gamma = \sqrt{mb}$ with b denoting the binding energy of the deuteron.

The analyticity region in $\cos \theta$ is, therefore, a cut plane with poles and branch points as shown in Fig. 1. Finally, we remark that our extrapolation

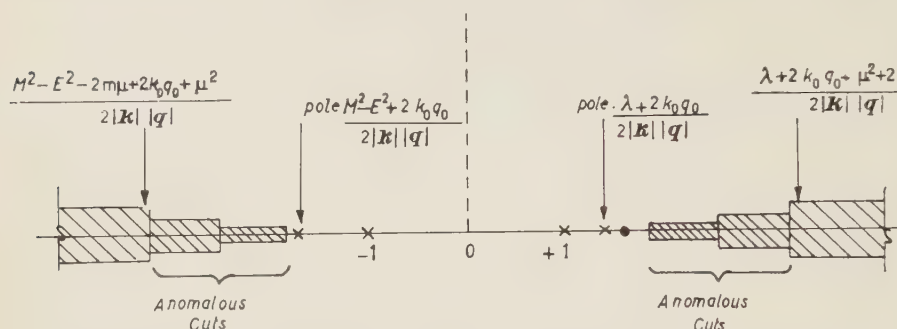


Fig. 1. - Singularities in $\cos \theta$ plane. The right-hand pole corresponds to that at $\Delta^2 = -m^2$.

procedure is based on the pole at $\Delta^2 = -m^2$. The other pole, coming from the crossed diagram, corresponds to the proton form-factor and does not interest us here.

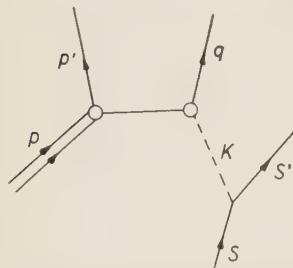


Fig. 2. - Diagram containing the pole.

In order to determine the neutron form-factor from the extrapolated cross-section, one needs a formula which exhibits the relationship between these two quantities. This is done by computing the contribution to the cross-section from the pole-diagram 2.

The contribution of the pole-diagram 2 to the cross-section is given by

$$(1) \quad \frac{d\sigma}{dP} = \frac{1}{96\pi^5} \cdot \frac{1}{E_e E_d} \cdot \frac{1}{|\mathbf{v}_1 - \mathbf{v}_2|} \cdot \frac{\alpha F^2}{\lambda^2 m_e} \cdot \frac{1}{(\Delta^2 + m^2)^2} \cdot \{ -\lambda(p' \cdot q)[4A^2 m_e^2 - 2\lambda A^2 + B^2(m_e^2 + s \cdot s')] + B^2[2(k \cdot p')(\lambda m_e^2 - \lambda^2/4) + 2\lambda(q \cdot s)p'(s \cdot s') + \lambda^2(p' \cdot s) + \frac{1}{2}\lambda^2 m^2] - m^2 B^2[2\lambda m_e^2 - \lambda^2] \},$$

where $|\mathbf{v}_1 - \mathbf{v}_2|$ is the relative velocity of deuteron and initial electron, E_e and E_d the initial particle energies and m_e the electron mass. α is the fine structure constant. The terms A and B occurring in (1) are given by

$$A = -\Gamma(1 + \Delta), \quad B = \frac{3m^2}{\gamma^2} \Delta \Gamma$$

with

$$\Gamma = \left[\frac{8\pi\gamma/m}{(1 + 2\Delta^2)(1 - e\gamma)} \right]^{\frac{1}{2}},$$

$$\Delta = (1 - e\gamma) Q\gamma^2, \quad \gamma = \sqrt{mb}.$$

In the above, b is the binding energy and Q the quadrupole moment of the deuteron, q is the neutron-proton triplet effective range, and F is the magnetic form-factor of the neutron obtained by writing the photon-neutron vertex as $\langle q | J_\mu | q' \rangle = F_1 \gamma_\mu + (q - q')_\nu \sigma_{\mu\nu} F_2$ we have put $F_1 = 0$ consistent with electron scattering experiments and written F for F_2 ; In obtaining (1) we needed the deuteron-neutron-proton vertex, which has been calculated by SAKITA (7) in connection with the problem of the photo-disintegration of the deuteron. We have used his results for this vertex. The factors A and B in (1) arise from this vertex. dP is the phase-space factor

$$\frac{1}{8} \frac{d^3 p'}{p_0'} \frac{d^3 q}{s_0'} \frac{d^3 s'}{q_0'} \delta^4(p' + s' + q - p - s),$$

one finds for this

$$dP = \frac{\pi}{4} \frac{d\lambda}{(q \cdot s')} \frac{dA^2}{\varepsilon} \frac{|\mathbf{p}'|}{4m} d\Omega_p,$$

where $d\Omega_p$ refers to the outgoing proton and ε and ε' are the initial and final electron energies respectively.

Finally, we have carried out summation over final particle spins and averaging over the initial particle polarizations in (1). From (1), we can write down the extrapolation formula:

$$(2) \quad (A^2 + m^2)^2 \frac{d\sigma}{dP} \bigg|_{A^2 = -m^2} = \frac{1}{96\pi^5} \cdot \frac{1}{E_e E_d} \frac{1}{|\mathbf{v} - \mathbf{v}_2|} \frac{\alpha F^2}{\lambda^2 m_e} \cdot \\ \cdot \left\{ -\lambda(p' \cdot q) [4m_e^2 A^2 - 2\lambda A^2 + B^2(m_e^2 + (s \cdot s'))] - m^2 B^2 [2\lambda m_e^2 - \lambda^2] + \right. \\ \left. + B^2 [2(k \cdot p')(\lambda m_e^2 - \lambda^2/4) + 2\lambda(q \cdot s)p' \cdot (s + s') + \lambda^2(p' \cdot s) + \frac{1}{2}\lambda^2 m^2] \right\} \bigg|_{A^2 = -m^2}.$$

In order to utilize (2), it will be necessary to measure the cross-section in terms of at least two variables. The electro-disintegration problem has five variables. These may be taken as A^2 , λ , E^2 , T_L , the laboratory kinetic energy of the incident electron and Φ , defined in the $\mathbf{p}' + \mathbf{q} = 0$ system as

$$\cos \Phi = \frac{(\mathbf{k} \times \mathbf{s}) \cdot (\mathbf{k} \times \mathbf{q})}{|(\mathbf{k} \times \mathbf{s}) \cdot (\mathbf{k} \times \mathbf{q})|}.$$

The cross-section may be measured as a function of A^2 and λ , the other three

(7) B. SAKITA: *Ph. D. Thesis at University of Rochester* (1958), unpublished.

variables having been integrated out. The extrapolation can then be carried out in terms of Δ^2 keeping λ fixed. The resulting form-factor will be a function of λ . One can, in this case, give a rough estimate of the experimental conditions for which the precedent extrapolation procedure is most favourable. Towards this end, we transform our variables to the laboratory system

$$(3) \quad \begin{cases} \Delta^2 = M(2E_{2L} - M) - m^2, \\ \lambda = 2T_L \varepsilon_{2L}(1 - \cos \psi), \end{cases}$$

where E_{2L} is the total energy of the final proton in the laboratory system, ε_{2L} the final electron kinetic energy and ψ the angle between the initial and final electrons in the same system. The electron mass has been neglected in obtaining (3). The relations (3) show that the extrapolation distance is $M(2E_{2L} - M)$. One must concentrate on small values of this expression since the error associated with extrapolation evidently increases with the distance to be extrapolated. The minimum value of the above expression is obtained by taking $E_{2L} = m$ i.e. when the final proton is at rest; $M(2E_{2L} - M)$ then just equals Mb . Experimentally, therefore, one has to measure the cross-section for a range of values of E_{2L} , concentrating on low final proton energies. The choice of low final proton energy has additional advantage. For scattering with large momentum transfer between electrons, low final proton energy implies correspondingly high energy for the final neutron. In this region the interaction between the final nucleons is unimportant and as such, the reliability of the proposed extrapolation procedure is improved.

Alternately, one could measure the cross-section in terms of all the five variables. In performing the extrapolation one would then keep all variables except one fixed.

Finally, we would like to remark that the present method is independent of the uncertainties regarding the choice of deuteron wave-functions and hence superior to methods that utilize the latter.

* * *

It is a pleasure to thank Professor R. E. MARSHAK for suggesting the problem and for stimulating discussions and encouragement. The author is grateful to Professor E. C. G. SUDARSHAN, Dr. I. H. BIRULA and Mr. H. J. SCHNITZER for helpful discussions. Lastly, the author's deep indebtedness goes to Professor C. J. GOEBEL for much patient advice and help throughout the course of this work.

APPENDIX

In this Appendix we locate the anomalous threshold singularities that appear in the electro-disintegration amplitude. A detailed study of these singu-

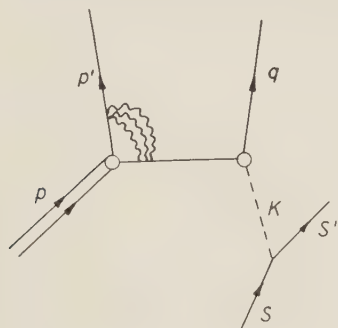


Fig. 3. - Diagrams giving rise to anomalous threshold branch points in electro-disintegration.

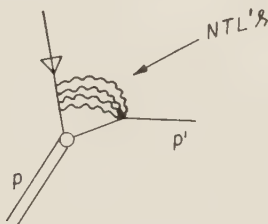


Fig. 4. - Diagram giving anomalous branch point in deuteron-proton-neutron vertex.

larities has been made by GOEBEL and SAKITA in their investigation of the deuteron-proton-neutron vertex⁽⁸⁾, and this Appendix is based on their work. The existence of the anomalous threshold singularities can be demonstrated in perturbation theory and their position located following Landau's⁽⁹⁾ method. The singularities in question arise from the class of diagrams in Fig. 3.

The wavy line represents a pion. To locate the singularities consider the corresponding deuteron-neutron-proton vertex (Fig. 4) with one nucleon (neutron) off the mass-shell and N pions exchanged between nucleons.

It's dual diagram is (Fig. 5).

p_c in Fig. 5 is the nucleon mass at the anomalous branch point. One obtains from Fig. 5

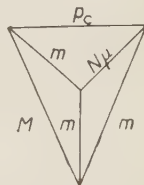


Fig. 5. - Diagram dual to diagram 4.

$$p_c^2 = m^2 + \frac{M^2}{2m^2} N^2 \mu^2 + \frac{NM\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - \frac{N^2 \mu^2}{4m^2}}.$$

In terms of A^2 of electro-disintegration problem, the above branch points

⁽⁸⁾ Private communication from Professor GOEBEL.

⁽⁹⁾ L. D. LANDAU: *Nuclear Physics*, **13**, 181 (1959). See also R. KARPLUS, C. M. SOMMERFIELD and E. H. WICHMANN: *Phys. Rev.*, **111**, 1187 (1958); J. C. TAYLOR: *Phys. Rev.*, **117**, 261 (1960).

occur at

$$\Delta^2 = -m^2 - \frac{M^2 N^2 \mu^2}{2m^2} - \frac{MN\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - N^2 \mu^2 / 4m^2}.$$

The first branch point occurs at ($N=1$)

$$(1a) \quad \Delta^2 = -m^2 - \frac{M^2}{2m^2} \mu^2 - \frac{M\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - \frac{\mu^2}{4m^2}} \simeq \\ \simeq -m^2 - 2\mu^2 - 4\mu\gamma + 0\left(\frac{1}{m^2}\right).$$

Similarly one finds that the anomalous branch points occurring through the crossed diagram occur at

$$\Delta^2 = E^2 + \lambda - M^2 - m^2 + \frac{M^2 N^2 \mu^2}{2m^2} + \frac{MN\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - \frac{N^2 \mu^2}{4m^2}},$$

with the first branch point occurring at

$$(1b) \quad \Delta^2 = E^2 + \lambda - M^2 - m^2 + \frac{M^2 \mu^2}{2m^2} + \frac{M\mu}{m} \sqrt{4m^2 - M^2} \sqrt{1 - \frac{\mu^2}{4m^2}} \simeq \\ \simeq E^2 + \lambda - M^2 - m^2 + 2\mu^2 + 4\mu\gamma + 0\left(\frac{1}{m^2}\right).$$

The singularities (1a) and (1b) respectively occur, in the $\cos \theta$ plane at

$$\cos \theta = \frac{\lambda + 2\mu^2 + 4\gamma\mu}{2|\mathbf{k}||\mathbf{q}|}, \\ \cos \theta = \frac{M^2 - 2\mu^2 - 4\gamma\mu - E^2 + 2k_0 q_0}{2|\mathbf{k}||\mathbf{q}|}$$

as stated in Fig. 1.

RIASSUNTO (*)

Si suggerisce un metodo di estrapolazione per determinare il fattore di forma magnetico del neutrone della elettrodisintegrazione del deutone.

(*) Traduzione a cura della Redazione.

Electroproduction of π -Mesons (*).

R. BLANKENBECLER

Princeton University - Princeton, N. J.

S. GARTENHAUS

Purdue University - Lafayette, Ind.

R. HUFF and Y. NAMBU

University of Chicago - Chicago, Ill.

(ricevuto l'11 Giugno 1960)

Summary. — An approximate evaluation is made of the dispersion relations for the production of pions in electron-nucleon collisions. The results are applicable at low energies in the final pion-nucleon barycentric system where the assumption that the (3, 3) state dominates the dispersion integrals is expected to be valid. Effects due to nuclear recoil and crossing symmetry are treated exactly to all orders.

1. — Introduction.

By use of the method of dispersion-relations, FUBINI, NAMBU and WATAGHIN ⁽¹⁾ have shown that experiments on the production of π -mesons in electron-nucleon collisions may be used as a tool for further study of the electromagnetic-structure of the nucleon. Unlike the case of elastic electron-nucleon scattering ⁽²⁾, for which the scattering amplitude is very simply related to the

(*) Supported in part by Air Force of Scientific Research and the Atomic Energy Commission.

⁽¹⁾ S. FUBINI, Y. NAMBU and V. WATAGHIN: *Phys. Rev.*, **111**, 329 (1958). Hereafter referred to as F.N.W.

⁽²⁾ G. HOFSTADTER: *Rev. Mod. Phys.*, **28**, 214 (1956). See also other papers quoted in this article.

form factors, in the meson-production problem, the amplitude depends on the nucleon structure in a fairly complicated way. This dependence can only be obtained by an evaluation of the dispersion-relations together with the unitarity condition. In the present note we present the results of such an approximate evaluation.

We shall need to assume only that the $(3, 3)$ pion-nucleon state dominates the dispersion integrals and further that states of more than one meson play no role. This means that the results obtained here are confined to experiments in which the center-of-mass energies in the final pion-nucleon state are near and below the resonance. No assumptions concerning the effects of nuclear recoils need be made. Now, for the case of photo-meson production at low energies ⁽³⁾ an expansion of the amplitude in inverse powers of the nucleon mass may be presumed to converge rapidly since only the photon and meson energies, k and ω respectively, are available for constructing a dimensionless expansion parameter ⁽⁴⁾. On the other hand, for the case of electro-pion production, there is an additional quantity available, namely the square of the invariant momentum transfer given up by the electron λ^2 , and this allows the construction of other dimensionless ratios; for example, $\lambda^2/M\omega$. Even though one is at reasonably low energies in the pion-nucleon system, in order to map out the form factors, one would like to have the freedom to allow values of $(\lambda^2)^{\frac{1}{2}}$ comparable to and even greater than a nucleon mass. Thus, since quantities such as $\lambda^2/M\omega$ make expansions in powers of $1/M$ very inconvenient, it is of importance to be able to evaluate the dispersion relations without making this expansion.

Following the methods given in a previous paper ⁽⁵⁾, we shall write down an approximate solution for the dispersion relations. Since the underlying philosophy and methods on electro-pion production already exist ^(1,6), and since the evaluational techniques have also been discussed ⁽⁵⁾, we shall give essentially only the final results with a minimum of subsidiary discussion.

2. - Solution of the dispersion relations.

We now proceed to write down the lowest order solution to the dispersion-relation as derived in reference ⁽⁵⁾. More precisely, it will be assumed that

⁽³⁾ G. F. CHEW, M. L. GOLDBERGER, F. E. LOW and Y. NAMBU: *Phys. Rev.*, **106**, 1345 (1957).

⁽⁴⁾ However, the arguments of CHARAP and FUBINI (*Nuovo Cimento*, **14**, 540 (1959)) even cast doubts on the validity of this expansion in this case.

⁽⁵⁾ R. BLANKENBECHER and S. GARTENHAUS: *Phys. Rev.*, **116**, 1297 (1959).

⁽⁶⁾ R. M. DALITZ and D. R. YENNIE: *Phys. Rev.*, **105**, 1598 (1957); R. B. CURTIS: *Phys. Rev.*, **104**, 211 (1956).

the phase of the difference between the total amplitude and the Born term less their (3, 3) projection, is given by the (3, 3) pion-nucleon scattering phase shift. The approximate solution then follows directly.

In the following discussion, we shall adhere as closely as possible to the notation of F.N.W. Thus, let p_1, p_2 be the initial and final momenta of the nucleon, q that of the meson, and k the momentum transfer from the electron. Out of these four-vectors, we can construct three invariants and these are chosen to be

$$(1) \quad \nu = -\frac{P \cdot k}{M}; \quad \nu_B = \frac{q \cdot k}{2M}; \quad \lambda^2 = k^2,$$

where P is the arithmetic average of the initial and final nucleon momenta. Other invariants involving the Dirac- γ matrices can also be constructed and the total amplitude M may be written in the form

$$(2) \quad M = A M_A + B M_B + \dots + F M_F,$$

where the six functions A, B, \dots, F depend on the three invariants of eq. (1) and M_A, M_B, \dots, M_F are defined by eq. (4) in F.N.W. For example, M_A is given by

$$M_A = \frac{i\gamma_5}{2} (\gamma \cdot \varepsilon \gamma \cdot k - \gamma \cdot k \gamma \cdot \varepsilon),$$

where ε_μ is the current vector for the electron, which on the assumption that the effects of the electron's structure may be neglected, is proportional to γ_μ taken between electron spinors. As in photoproduction, A, B, \dots, F are matrices in isotopic spin space. The assumption of charge independence allows only three independent functions for each partial amplitude, and these are written as $A^+, A^-, A^0, \dots, F^0$ where the symbols (\pm) have the meaning as given in F.N.W. Thus, altogether there are eighteen functions to consider and we shall designate the i -th of these by H_i . The dispersion relations are given by (1)

$$(3) \quad H_i(\nu, \nu_B, \lambda^2) = B_i(\nu, \nu_B, \lambda^2) + \frac{1}{\pi} \int_{\nu_0}^{\infty} d\nu' \operatorname{Im} H_i(\nu', \nu_B, \lambda^2) \left[\frac{1}{\nu' - \nu - i\varepsilon} \pm \frac{1}{\nu' \pm \nu} \right],$$

where $\nu_0 = 1 + (1/2M) + \nu_B$, and where $A^{+,0}, B^{+,0}, C^-, D^\pm, E^-, F^-$ are even functions of ν and the remaining nine are odd. The functions $B_i(\nu, \nu_B, \lambda^2)$ will be called the Born terms and are explicitly given by equations (8), (8') and (10) of F.N.W. (7). The nucleon structure factors are functions only of λ^2 and are explicitly contained in the Born terms.

(7) There is a misprint in eq. (10) and C_E^- as written there should be multiplied by a minus sign.

Now in order to write down the approximate solution to eq. (3) by use of the methods of reference (5) it is convenient to change variables from ν to x , where x is given by

$$x' = \nu - \nu_B = \frac{W^2 - M^2}{2M}$$

in which W is the total meson-nucleon energy in their center of mass frame. In terms of this variable, the approximate solution of eq. (3) is given by

$$(4) \quad H_j(x, \nu_B, \lambda^2) = B_j(x, \nu_B, \lambda^2) + \frac{\exp[i\delta(x)]}{\pi} \int_{1+\frac{1}{2}M}^{\infty} dy \sin \delta(y) b_j(y, \nu_B, \lambda^2) \cdot \exp[\Delta(y, x, \nu_B)] \left[\frac{1}{y-x-i\epsilon} \pm \frac{1}{y+x+2\nu_B} \right],$$

where $\delta(x)$ is the (3, 3) phase shift and $\Delta(x', x, \nu_B)$ is given by

$$(5) \quad \Delta(x', x, \nu_B) = \varrho(x, \nu_B) - \varrho(x', \nu_B)$$

and where finally, ϱ is given by

$$(6) \quad \varrho(x, \nu_B) = \frac{P}{\pi} \int_{1+\frac{1}{2}M}^{\infty} dy \delta(y) \left[\frac{1}{y-x} + \frac{1}{y+x+2\nu_B} \right].$$

In general, the integral in eq. (6) will not converge unless $\delta \rightarrow 0$ at infinity; but as has been shown (6), one subtraction in ϱ can be made without adding a new constant to Δ of eq. (5). In this sense, eq. (6) is valid even if δ approaches a non-zero constant for large energies. Finally, the eighteen functions $b_j(y, \nu_B, \lambda^2)$ are the (3, 3) projections of the Born terms and their determination is our next task. We note that the solution given by eq. (4) has all of the symmetry in ν and all of the analyticity properties in both ν and ν_B as is manifest in eq. (3). The ambiguities have been removed in accordance with the discussion in reference (1). Further the amplitude is proportional to the pion-nucleon coupling constant f , and depends linearly on the form-factors themselves. In this respect, at least, the solution is very similar to the amplitude for elastic electron-nucleon scattering.

It should also be pointed out that this solution may be described in terms of the language associated with the Mandelstam representation. The solution in eq. (4) for the (3, 3) state satisfies unitarity on the positive energy rescattering cut and is chosen to have the negative energy cut coming essentially from the Born-approximation with a correction due to the crossing symmetry.

(8) S. GARTENHAUS and R. BLANKENBECLER: *Phys. Rev.*, **116**, 1305 (1959).

In this sense it is close to the work of FRAZER and FULCO ⁽⁹⁾ on the annihilation of a nucleon-antinucleon pair into two pions.

In order to obtain the b_j , it is convenient to reduce the four-component Dirac-matrices in eq. (2) to two component ones and to express all quantities in the pion-nucleon center of mass frame. We define six functions, $\mathcal{F}_1, \mathcal{F}_2, \dots, \mathcal{F}_6$, and six combinations involving two component Pauli-spin matrices, $\Sigma_1, \Sigma_2, \dots, \Sigma_6$, such that the amplitude of eq. (2) can be written in the form

$$(7) \quad M = \sum_{i=1}^6 \Sigma_i \mathcal{F}_i,$$

where the Σ_i are defined by

$$(8) \quad \begin{cases} \Sigma_1 = i\boldsymbol{\sigma} \cdot \mathbf{a}; & qk\Sigma_2 = \boldsymbol{\sigma} \cdot \mathbf{q} \boldsymbol{\sigma} \cdot (\mathbf{k} \times \mathbf{a}); & qk\Sigma_3 = i\boldsymbol{\sigma} \cdot \mathbf{k} \mathbf{q} \cdot \mathbf{a}; \\ q^2\Sigma_4 = i\boldsymbol{\sigma} \cdot \mathbf{q} \mathbf{q} \cdot \mathbf{a}; & k^2\Sigma_5 = i\boldsymbol{\sigma} \cdot \mathbf{k} \mathbf{k} \cdot \mathbf{a}; & qk\Sigma_6 = i\boldsymbol{\sigma} \cdot \mathbf{q} \mathbf{k} \cdot \mathbf{a}. \end{cases}$$

In these relations, all three component vectors are evaluated in the center-of-mass system and \mathbf{a} is given by

$$\mathbf{a} = \boldsymbol{\varepsilon} - \frac{\mathbf{k} \cdot \boldsymbol{\varepsilon}}{k_0^2} \mathbf{k},$$

where we have used gauge invariance but have not restricted ourselves to a particular gauge. The relations between the \mathcal{F}_i and A, B, \dots , is readily obtained from equations (2), (7) and (8) and the result is

$$(9) \quad \begin{cases} F_1 = \frac{2M}{\omega O_1} \mathcal{F}_1 = A - \frac{2Mv_B}{\omega} (C - D) + \omega D + \frac{\lambda^2}{\omega} F, \\ F_2 = \frac{2M(E_1 + M)}{qkO_2(W + M)} \mathcal{F}_2 = -A - \frac{2Mv_B}{W + M} (C - D) + (W + M)D + \frac{\lambda^2}{W + M} F, \\ F_3 = \frac{2M(E_1 + M)}{qkO_1(W + M)} \mathcal{F}_3 = \omega B + C - D - \frac{\lambda^2}{W + M} E, \\ F_4 = \frac{2M}{q^2O_2\omega} \mathcal{F}_4 = -(W + M)B + C - D + \frac{\lambda^2}{\omega} E, \\ F_5 = \frac{2M(E_1 + M)}{k^2O_1} \mathcal{F}_5 = -A - 2Mv_B B - \omega D + 2Mv_B E - (W + M)F, \\ F_6 = \frac{2M}{qkO_2} \mathcal{F}_6 = -\frac{k_0}{E_1 + M} A + 2Mv_B B - \frac{2Mv_B}{E_1 + M} (C - D) + \\ \quad + k_0 \frac{E_1 + M}{W + M} D - 2Mv_B E - k_0 \frac{W + M}{E_1 + M} F, \end{cases}$$

⁽⁹⁾ W. FRAZER and J. FULCO, and University of California at Berkeley, Preprint U.C.R.L. 8806.

where E_1 and E_2 are the initial and final nucleon energies, $\omega = W - M$, k_0 is the time component of k , and O_1 and O_2 are given by

$$O_1 = \sqrt{(E_1 + M)(E_2 + M)}; \quad O_2 = \sqrt{\frac{E_1 + M}{E_2 + M}}.$$

In particular, the Born terms may be grouped as a row matrix, according to eq. (7) and we find, by neglecting those parts which make no contribution to the (3, 3) state, the result

$$(10) \quad \begin{cases} F_\mu^B = \frac{-2Mf\mu_v}{kq} \frac{1}{\cos\theta + a} \left(M, -M, 1, 1, \frac{\omega}{2}, \frac{(W+M)(E_1-M)}{2(E_1-M)} \right), \\ F_e^B = \frac{e^v f}{kq} \frac{1}{\cos\theta + a} \left(0, 0, \frac{\omega}{W+M}, \frac{M+W}{\omega}, \frac{\omega}{2}, \frac{W+M}{2} \right), \\ F_\pi^B = \frac{ef}{kq} \frac{1}{\cos\theta - b} \left(0, 0, \frac{2M}{M+M}, \frac{-2M}{\omega}, -M, M \right), \end{cases}$$

where we have divided the result into the parts coming from the magnetic moment, the charge, and the contributions from the meson-current diagram. In eq. (10), θ is the angle between the momenta of the virtual photon and the final meson, $\mu_v(\lambda^2)$ is the total vector part of the magnetic moment. In terms of form factors it is given by

$$\mu_v = \mu'_P F_2^P(\lambda^2) - \mu_N F_2^N(\lambda^2) + \frac{e^v}{2M},$$

where $\mu'_{p(N)}$ is the anomalous part of the magnetic-moment of the proton (neutron) and e^v is given by

$$e^v = e[F_1^P(\lambda^2) - F_1^N(\lambda^2)]$$

and where $F_1^P, F_2^P, F_1^N, F_2^N$ are as defined by HOFSTADTER⁽²⁾. The quantities a and b are given by

$$(11) \quad \begin{cases} a = \frac{2k_0 E_2 + \lambda^2}{2qk}, \\ b = \frac{2k_0 q_0 + \lambda^2}{2qk}, \end{cases}$$

which in the limit of $\lambda^2 \rightarrow 0$ approach the inverse of the final nucleon and meson velocity respectively. The four center-of-mass energies E_1, E_2, q_0 and k_0 may be expressed in terms of the invariants W^2 and λ^2 and are given by the

relations

$$E_1 = \frac{W^2 + M^2 + \lambda^2}{2W},$$

$$E_2 = \frac{W^2 + M^2 - 1}{2W},$$

$$q_0 = \frac{W^2 - M^2 + 1}{2W},$$

$$k_0 = \frac{W^2 - M^2 - \lambda^2}{2W}.$$

In terms of the initial and final electron momenta in the laboratory the invariants W^2 and λ^2 may be written

$$W^2 = M^2 - \lambda^2 + 2M(\varepsilon_1 - \varepsilon_2),$$

$$\lambda^2 = 2[\varepsilon_1 \varepsilon_2 - l_1 l_2 \cos \varphi - m_e^2] \simeq 2\varepsilon_1 \varepsilon_2 (1 - \cos \varphi),$$

where ε_1 , ε_2 are the initial and final electron energies in the laboratory, l_1 , l_2 the magnitudes of the corresponding momenta and φ the angle of scattering in the laboratory. It is easy to show, that the denominators in eq. (10) never vanish for real angles and thus the projections can be made without difficulty.

Finally, therefore, we substitute the Born terms given by eq. (10) into eq. (7), project the $P_{\frac{3}{2}}$ state out of this expression and regroup the results in the form needed for eq. (7). For the magnetic-moment terms the results are

$$\frac{F_{\mu}^1}{A_{\mu}} = 3M\bar{\alpha} - \frac{3}{2} \frac{Mqk(W+M)}{\omega(E_1+M)(E_2+M)} \bar{\gamma} + \frac{q^2}{E_2+M} \left(1 - \frac{3}{2} a\bar{\beta}\right),$$

$$\frac{F_{\mu}^2}{A_{\mu}} = \frac{M(E_1+M)(E_2+M)}{qk(W+M)} \omega\bar{\alpha} - \frac{M}{2} \bar{\gamma} - \frac{(E_2+M)\bar{\beta}}{2},$$

$$\frac{F_{\mu}^3}{A_{\mu}} = \frac{3}{2} \bar{\beta} + \left(1 - \frac{3}{2} a\bar{\beta}\right) \left(\frac{E_1+M}{E_2+M}\right) \left(\frac{\omega}{W+M}\right) \frac{q}{k},$$

$$F_{\mu}^4 = 0,$$

$$\frac{F_{\mu}^5}{A_{\mu}} = \frac{q^2}{k^2} \frac{\omega(E_1+M)}{E_2+M} \left(\frac{5}{2} a\bar{\gamma} - 2\bar{\alpha}\right) + \frac{3}{2} \omega\bar{\alpha} - \frac{3}{4} \frac{q(W+M)}{k(E_2+M)} (2E_2 + E_1 - M) \bar{\gamma},$$

$$\begin{aligned} \frac{F_{\mu}^6}{A_{\mu}} = & \left(\frac{(W+M)(2E_2+E_1+M)}{4(E_1+M)} - \frac{\omega a}{2} \frac{q}{k} \right) \bar{\gamma} - \\ & - \frac{\omega(E_1+M)(E_2+M)}{2qk} \bar{\alpha} - \frac{(W+M)(E_2+M)}{2(E_1+M)} \bar{\gamma}. \end{aligned}$$

In these expressions $\bar{\alpha}$, $\bar{\beta}$, $\bar{\gamma}$ are defined by the relations

$$(13) \quad \left\{ \begin{aligned} \bar{\alpha} &= 1 - \frac{a}{2} \ln \frac{a+1}{a-1}, \\ \bar{\beta} &= a + \frac{1-a^2}{2} \ln \frac{a+1}{a-1}, \\ \bar{\gamma} &= 3a + \frac{1-3a^2}{2} \ln \frac{a+1}{a-1} = 3\bar{\beta} + \frac{2}{a}\bar{\alpha} - \frac{2}{a}. \end{aligned} \right.$$

where a is given by eq. (11). The quantity A_μ is given by $A_\mu = -(2Mf\mu_r/qk)$ and x is the cosine of the angle between \mathbf{q} and \mathbf{k} . The contributions of the charge may be similarly grouped and the result is

$$(14) \quad \left\{ \begin{aligned} \frac{F_e^1}{xB_e} &= \frac{q^2}{E_2+M} \frac{W+M}{W-M} \left(1 - \frac{3}{2} a \bar{\beta} \right), \\ \frac{F_e^2}{B_e} &= -\frac{1}{2} \frac{W-M}{W+M} (E_2+M) \bar{\beta}, \\ \frac{F_e^3}{B_e} &= \frac{3}{2} \frac{W-M}{W+M} \bar{\beta} + \frac{W+M}{W-M} \frac{qk}{(E_2+M)(k_0+(\lambda^2/\omega))} \left(1 - \frac{3}{2} a \bar{\beta} \right), \\ F_e^4 &= 0, \\ \frac{F_e^5}{xB_e} &= \frac{q^2}{k^2} \frac{(E_1+M)(W+M)}{E_2+M} \left(\frac{5}{2} a \bar{\gamma} - 2\bar{\alpha} \right) + \frac{3}{2} \omega \bar{\alpha} - \\ &\quad - \frac{3}{4} \frac{q}{k} \frac{(W+M)(E_1+M)}{E_2+M} \bar{\gamma} - \frac{3}{2} \omega \frac{q}{k} \bar{\gamma}, \\ \frac{F_e^6}{B_e} &= \left[\frac{\omega}{2} \frac{(E_2+M)}{(E_1+M)} + \frac{W+M}{4} - \frac{q}{2k} (W+M) \alpha \right] \bar{\gamma} - \\ &\quad - \frac{k}{2q} \frac{\omega(E_2+M)}{E_1+M} \bar{\alpha} - \frac{\omega}{2} \frac{(E_2+M)}{(E_1+M)} \bar{\beta}, \end{aligned} \right.$$

where B_e is given by

$$B_e = \frac{f e^v}{qk}.$$

Finally, from the meson current diagram we obtain

$$(15) \quad \left\{ \begin{aligned} \frac{F_\pi^1}{xB_\pi} &= -\frac{2M}{\omega} \frac{\mathbf{q}^2}{E_2 + M} \left(1 - \frac{3}{2} b \hat{\beta}\right), \\ \frac{F_\pi^2}{B_\pi} &= \frac{M(E_2 + M)}{W + M} \hat{\beta}, \\ \frac{F_\pi^3}{B_\pi} &= -\frac{3M}{W + M} \hat{\beta} - \frac{2M}{\omega} \frac{qk}{E_2 + M} \frac{1}{k_0 + \gamma^2/\omega} \left(1 - \frac{3}{2} b \hat{\beta}\right), \\ F_\pi^4 &= 0, \\ \frac{F_\pi^5}{xB_\pi} &= -2M \frac{\mathbf{q}^2}{k^2} \left(\frac{E_1 + M}{E_2 + M}\right) \left(\frac{5}{2} b \hat{\gamma} - 2\hat{\alpha}\right) - 3M\hat{\alpha} + 3M \frac{q}{k} \hat{\gamma} + \\ &\quad + \frac{3}{2} M \frac{q}{k} \frac{E_1 + M}{E_2 + M} \hat{\gamma}, \\ \frac{F_\pi^6}{B_\pi} &= \left[M \frac{q}{k} b - \frac{M}{2} - M \frac{(E_2 + M)}{E_1 + M} \right] \hat{\gamma} + M \frac{E_2 + M}{E_1 + M} \hat{\beta} + M \frac{k}{q} \frac{E_2 + M}{E_1 + M} \hat{\alpha}, \end{aligned} \right.$$

where B_π is given by $B_\pi = ef/qk$ and e contains all of the structure in the meson propagator and that occurring at the pion vertex where the virtual photon is absorbed. The quantities $\hat{\alpha}$, $\hat{\beta}$, $\hat{\gamma}$ are defined by expressions identical to those in eq. (13) with a replaced by the quantity b .

To summarize, the procedure for obtaining the eighteen quantities b_j in the solution given by eq. (4) is as follows: Take eqs. (12), (14), (15) and substitute these on the left hand sides of eqs. (9). Then solve eq. (9) to obtain the solutions which we shall refer to as a , b , ..., f and are explicitly given by

$$\begin{aligned} a &= 2Md + \left[\frac{\omega}{2W} F_1 - \frac{W + M}{2W} F_2 \right], \\ b &= \frac{\lambda^2}{4WMk_0v_B} \left\{ \frac{\omega}{2M} F_1 + \frac{k_0(W + M)}{2W(E_1 + M)} F_2 + \frac{2Mv_B}{2W\lambda^2} (W^2 - M^2)(F_3 - F_4) + \right. \\ &\quad \left. + \frac{\omega}{2W} F_5 - \frac{W + M}{2W} F_6 \right\}, \\ c &= d + \frac{\omega}{2W} F_4 + \frac{W + M}{2W} F_3, \\ d &= \frac{1}{(2W)^2 k_0} \cdot \\ &\quad \cdot \left\{ (W^2 - M^2) F_1 + k_0 \frac{(W + M)^2}{E_1 + M} F_2 + 2Mv_B [\omega F_4 + (W + M) F_3] + \lambda^2 (F_5 + F_6) \right\}, \\ e &= \frac{W^2 - M^2}{\lambda^2} b + \frac{W^2 - M^2}{2W\lambda^2} (F_4 - F_3), \\ f &= -d + \frac{W + M}{2W(E_1 + M)} F_2 - \frac{F_5 + F_6}{2W}, \end{aligned}$$

where

$$F_1 = F_\mu^1 + F_e^1 + F_\pi^1, \quad \dots$$

In terms of these six functions, the eighteen quantities b_i in eq. (4) are as follows. The six quantities $a^{(+)}, b^{(+)}, \dots, f^{(+)}$ which go with the isotopic spin (+) sign are given by $\frac{2}{3}a, \frac{2}{3}b, \dots, \frac{2}{3}f$. The six quantities $a^{(-)}, b^{(-)}, \dots, f^{(-)}$ which go with the isotopic spin (-) sign are given by $-\frac{1}{3}a, -\frac{1}{3}b, \dots, -\frac{1}{3}f$. Lastly, the six quantities a^0, b^0, \dots, f^0 vanish identically and thus the complete amplitude for this particular isotopic spin dependence is given by the Born approximation itself.

3. - Concluding remarks.

Following the procedure outlined above, an approximate evaluation of the Dispersion Relations may be obtained. The advantages of the present results over previous ones are substantially that no expansion in inverse powers of the nucleon mass had to be made, and secondly, that the results are available in completely covariant form. The latter has the advantage that the longitudinal contributions to the matrix element will always remain finite even, if the time component of k approaches zero. Also, the results have been obtained without going into a particular gauge, and the freedom so afforded may allow for some further simplifications.

A final point of interest deals with the contributions to the Born terms of the meson current diagram (Fig. 1).

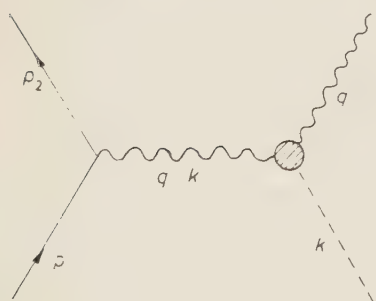


Fig. 1.

Following F.N.W., we have neglected the electromagnetic structure at the pion-photon vertex and have referred to this contribution by the symbol e . Actually, this quantity is a function of both $(q-k)^2 = (P_1 - P_2)^2$ and λ^2 and it is *not* what we would like to call the form factor for the pion. However, as has been shown by FRAZER⁽¹⁰⁾, in experiments in which one detects both the final electron and pion, that is one observes all three invariants, x , ν_B , and λ^2 , one may extrapolate the cross-section to the mass shell of the virtual pion in Fig. 1. This corresponds to the value $(q-k)^2 = -1$, or $\lambda^2 = +4M\nu_B$. At this unphysical angle, the quantity e in eq. (10) becomes precisely the electromagnetic stru-

⁽¹⁰⁾ W. FRAZER: *Phys. Rev.*, **115**, 1763 (1959).

eture factor for the pion. Thus, provided no experimental difficulties occur, and provided one may extrapolate, the experiments on electro-pion production allow a determination of the structure of the pion as well as the nucleon. A numerical evaluation of these results is now in progress in order to aid in the interpretation of the experiments of PANOFSKY and ALLTON ⁽¹¹⁾.

⁽¹¹⁾ W. K. H. PANOFSKY and E. A. ALLTON: *Phys. Rev.*, **110**, 1155 (1958). Further experiments are in progress. We wish to thank Prof. PANOFSKY for many interesting conversations concerning his experiments.

RIASSUNTO (*)

Si fa una valutazione approssimata delle relazioni di dispersione per la produzione di pioni nelle collisioni elettrone-nucleone. I risultati sono applicabili a basse energie al sistema baricentrico finale pione-nucleone, in cui ci si attende che sia valida l'ipotesi che lo stato $(3, 3)$ domina gli integrali di dispersione. Gli effetti dovuti al rinculo nucleare e alla simmetria incrociata vengono trattati esattamente in tutti gli ordini.

(*) Traduzione a cura della Redazione.

Interactions of 1.15 GeV/c K^- -Mesons in Emulsion. — I.

C. M. GARELLI, B. QUASSIATI and M. VIGONE

Istituto di Fisica dell'Università - Torino
Istituto Nazionale di Fisica Nucleare - Sezione di Torino

(ricevuto il 30 Giugno 1960)

Summary. — In this paper are reported the results of an analysis of 1675 interactions of high energy K^- -mesons with the emulsion nuclei. The main results concern the emission of charged hyperons, their energy and angular distribution, the ratio $(\Sigma^- \text{ emitted})/(\Sigma^+ \text{ emitted})$; an estimate is given of the probability of Σ inelastic reemission.

1. — Introduction.

The interaction of K^- mesons with hydrogen, deuterium and emulsion nuclei have been accurately analysed by several groups ⁽¹⁻⁸⁾ up to a K^- momentum of 400 MeV/c. At higher energy of the K^- -meson, the available data

(1) L. W. ALVAREZ, H. BRADMER, P. FALK-VAIRANT, J. D. GOW, A. H. ROSENFELD, F. T. SOLMITZ and R. D. TRIPP: *Nuovo Cimento*, **5**, 1026 (1957).

(2) F. C. GILBERT, C. E. VIOLET and R. S. WHITE: *Phys. Rev.*, **107**, 228 (1957).

(3) F. M. WEBB, E. L. ILOFF, F. H. FEATHERSON, W. W. CHUPP, G. GOLDBABER and S. GOLDBABER: *Nuovo Cimento*, **8**, 899 (1958).

(4) G. HORNBOSTEL and G. T. ZORN: *Phys. Rev.*, **109**, 165 (1958).

(5) K^- COLLABORATION: *Nuovo Cimento*, **14**, 315 (1959).

(6) Y. EISENBERG, W. KOCH, E. LOHRMANN, M. NIKOLIĆ, M. SCHNEEBERGER and H. WINZELER: *Nuovo Cimento*, **9**, 745 (1958).

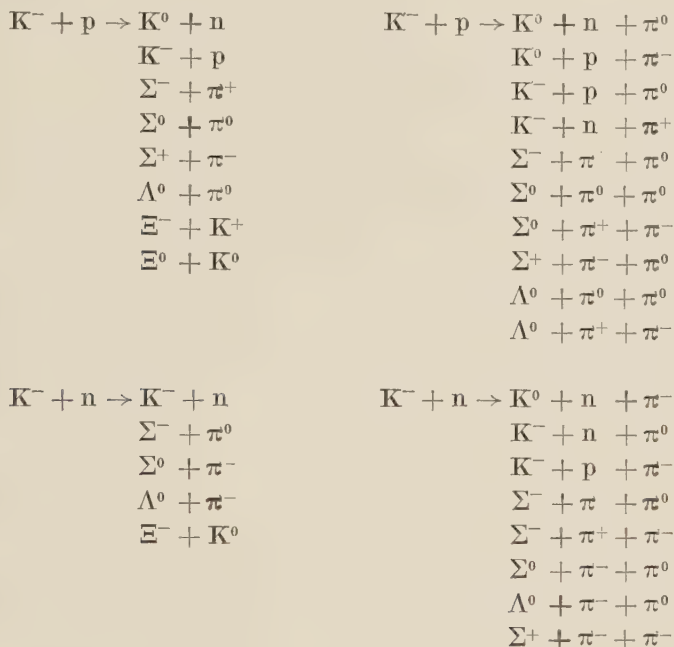
(7) N. HORWITZ, D. MILLER, J. MURRAY, M. SCHWARTZ, H. TAFT, O. DAHL, V. MONARA and P. WHITE: *Report at the Annual International Conference on High Energy Physics at Kiev* (1959).

(8) J. P. BERGE, W. HUMPHREY, P. NORDIN, A. H. ROSENFELD, R. ROSS, F. T. SOLMITZ and R. D. TRIPP: *Report at Annual International Conference on High Energy Physics at Kiev* (1959).

are only the very previous ones collected in the Report of the Conference on High Energy Physics, hold in Kiev in 1959 ⁽⁹⁾.

The purpose of the present work is to present the results obtained up to now ^(*) in this laboratory, on the interactions of 1.15 GeV/c K^- -mesons in emulsions.

The possible reactions with one nucleon are the following:



The production of more than two π -mesons as well as the interactions with more than one nucleon are possible at the considered K^- energy, but their percentage is certainly not very large.

In this paper we are mainly concerned with the emission of charged hyperons, their energy spectrum and their interactions with emulsion nuclei.

We hope to be able to give, in a subsequent paper an analysis of the K^- re-emissions and of the π -meson production. Now, we give only a rough evaluation of the percentage of K^- -mesons re-emitted and of the multiplicity of pions in the stars produced by high energy K^- -mesons.

⁽⁹⁾ L. W. ALVAREZ, P. EBERHARD, M. L. GOOD, W. GRAZIANO, H. K. TICHÖ and S. G. WOICICKI: *Report at the Annual International Conference on High Energy Physics at Kiev* (1959).

^(*) The very previous results given in a preceding paper (C. M. GARELLI, B. QUAS-
SIATI, L. TALLONE and M. VIGONE: *Nuovo Cimento*, **13**, 1294 (1959)) are also included
in the present work.

We point out that the scanning has been done in order to find the stars produced by K^- -mesons, so that nothing can be said about elastic scatterings.

2. - Exposure details and beam composition.

A stack of 140 sheets, (9 in. \times 6 in. \times 600 μ m), G-5 emulsions, was exposed to the 1.15 GeV/c K^- beam of the Berkeley bevatron.

The beam has been developed by GOOD and TICHO⁽¹⁰⁾ and the average composition is expected to be: 1.5 K^- ; 4.5 μ^- ; 0.2 π^- . The actual composition during our exposure is not known; we tried to check the ratio $\mu/(K+\pi)$ measuring the attenuation of the component producing stars in our stack. The details of the calculation are given in Appendix I and the results are the following: the component $(K+\pi)$ has a mean free path for star production of (29^{+9}_{-6}) cm, and its ratio to the μ component is $1/(2.4 \pm 0.8)$. It can be seen that this result is compatible with the value given by GOOD and TICHO within the statistical fluctuations.

In what follows we shall take, as an order of magnitude, that $\sim 10\%$ of the particles producing stars are π^- -mesons.

3. - Scanning method and experimental results.

A total of 81.2 cm³ has been explored looking for stars produced by the tracks of the beam. The total number of stars found in this way is 1577. To this number, 98 stars found by track following have been added. The scanning by track following gives unbiased results, while the scanning by area can give a loss of small stars. Actually this is not the case, as proved by the mean number of prongs per star that results to be:

5.6 ± 0.6 in the stars found by track following;

6.3 ± 0.15 in the stars found by area scanning.

All the prongs with ionization corresponding to a value of $\beta \leq 0.70$ have been followed until they came to rest, or interacted, or left the stack. The tracks that interacted or left the stack have been analysed in order to decide if they were due to a π -meson, a K -meson or to a particle of protonic mass.

⁽¹⁰⁾ L. L. ALVAREZ, P. EBERHARD, M. L. GOOD, W. GRAZIANO, H. K. TICHO and S. G. WOICICKI: *Phys. Rev. Lett.*, **2**, 215 (1957).

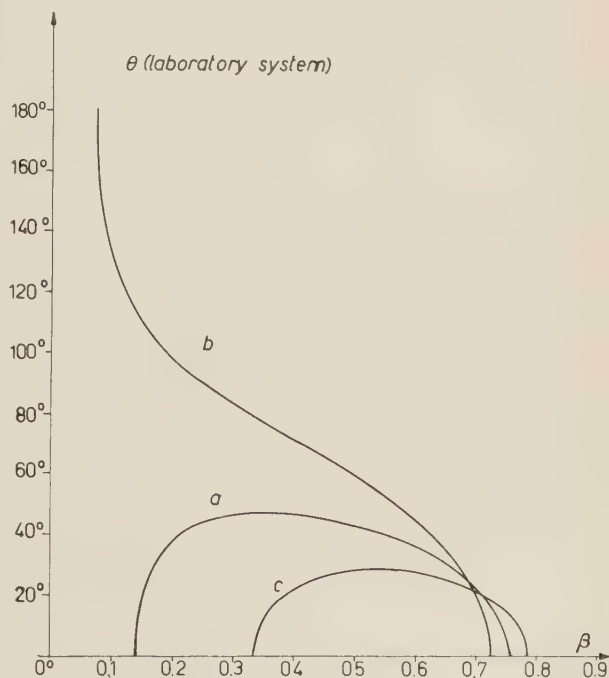
Moreover, in a cone of 45° around the primary direction, also the tracks with ionization corresponding to a value of $\beta > 0.70$ have been followed. In this way we can be sure that the tracks that have not been followed (*i.e.* the tracks of particles with $\beta > 0.70$ and emitted at an angle $> 45^\circ$ with the primary direction) are due to π or K -mesons, but certainly not to Σ -hyperons (see Fig. 1).

The results are the following:

particles emitted with $\beta \leq 0.70$	{	stable prongs	9 461
		charged π -mesons	240
		K^- reemitted	33
		charged Σ -hyperons	168
		hyperfragments	36
particles emitted with $\beta > 0.70$			628

No certain case of Ξ^- -hyperons was observed. All the 168 charged hyperons were carefully studied and in 8 cases only, due to the unfortunate geometrical conditions, a sure discrimination between Σ and Ξ -hyperons was not possible. This fact indicates a very small value for the cross-section of Ξ -hyperon production, and it is not in disagreement with the very few data available on this subject (⁹⁻¹¹).

Fig. 1. - Angle of emission of Σ hyperons versus the β value of the Σ hyperons emitted in the reaction $K + N \rightarrow \Sigma + \pi$: curve *a* corresponds to a reaction with a free nucleon; curves *b* and *c* correspond to a reaction with a bound nucleon, taking into account a Fermi momentum with, respectively, the opposite direction or the same direction of the incoming K -meson.



(¹¹) W. B. FOWLER, L. M. GOWELL and J. I. SHONLE: *Nuovo Cimento*, **11**, 438 (1959).

An unusual hyperon decay has been observed during this analysis and described in a separate paper (C. M. GARELLI, B. QUASSIATI and M. VIGONE: *Nuovo Cimento*, **16**, 960 (1960)).

4. - Emission of charged Σ hyperons.

The total number of charged Σ -hyperons, recognized between the prongs of the 1675 analysed stars, is 170. Two of them are emitted with $\beta > 0.70$. The observed events are the following:

Σ^- captured	at rest	21
$\Sigma^- \rightarrow \pi^-$	in flight	21
$\Sigma^+ \rightarrow \pi^+$	in flight	8
$\Sigma^+ \rightarrow p$	in flight	18
$\Sigma^+ \rightarrow p$	at rest	6
$\Sigma^\pm \rightarrow \pi^\pm$	in flight	94
Σ^\pm interacting	in flight	2

The number of Σ^- captured at rest includes only those negative hyperons that come to rest giving a capture star of at least two prongs.

The $\Sigma \rightarrow \pi$ events are recognized by the kinematics of the decay; we can give undoubtedly the charge of the hyperon only when the π -meson produced in the decay comes to rest in the emulsion.

The $\Sigma^- \rightarrow p$ decays at rest are certainly recognized from the range of the decay proton; the $\Sigma^+ \rightarrow p$ decays in flight are recognized from the kinematics of the event, but the interpretation can be doubtful in the cases where the β value of the decay proton is lower than the β value of the decaying hyperon.

The Σ interacting in flight have been recognized from the features of the produced stars (see Appendix III).

From the preceding considerations it follows that the numbers that need to be corrected for observation losses are the numbers of Σ^- captured at rest and the number of $\Sigma^+ \rightarrow p$ decays.

The evaluation of these corrections, and the criterium used to attribute the sign of the electric charge to the 94 cases of Σ -hyperons with undetermined charge, are given in Appendix II.

The results of these corrections give the most probable features of the Σ -hyperons emitted from the analysed stars, and are the following:

Σ^- captured at rest	75
$\Sigma^- \rightarrow \pi^-$	94
$\Sigma^+ \rightarrow \pi^+$	29
$\Sigma^+ \rightarrow p$	29

From the above given numbers, we can derive immediately the ratio $\Sigma_{\text{emitted}}^-/\Sigma_{\text{emitted}}^+$ that turns out to be: 2.9.

To the above estimated number of 227 Σ -hyperons, we must apply a correcting factor of 1.1 to take into account, as described in Section 7, the loss due to the fact that some of the Σ -hyperons interact in flight giving rise to stars undistinguishable from the stars produced by protons. So we obtain that the most probable number of charged Σ -hyperons emitted from the 1675 analysed stars is 250, of which:

- 186 are negatively charged Σ -hyperons, and
- 64 are positively charged Σ -hyperons.

5. - Energy and angular distributions of charged Σ hyperons.

We give in Fig. 2 the histogram of the energy distribution of the charged Σ -hyperons in the laboratory system. The dashed curve is the one obtained from the actually observed events. In the preceding section we described the corrections to be applied for various kinds of losses; only one of them, the one concerning the number of Σ^- -hyperons captured at rest, has an influence on the shape of the observed spectrum; the full line represents the energy spectrum obtained taking into account this correction.

The t curve has been calculated under the assumption that all charged Σ -hyperons are produced in the reaction:

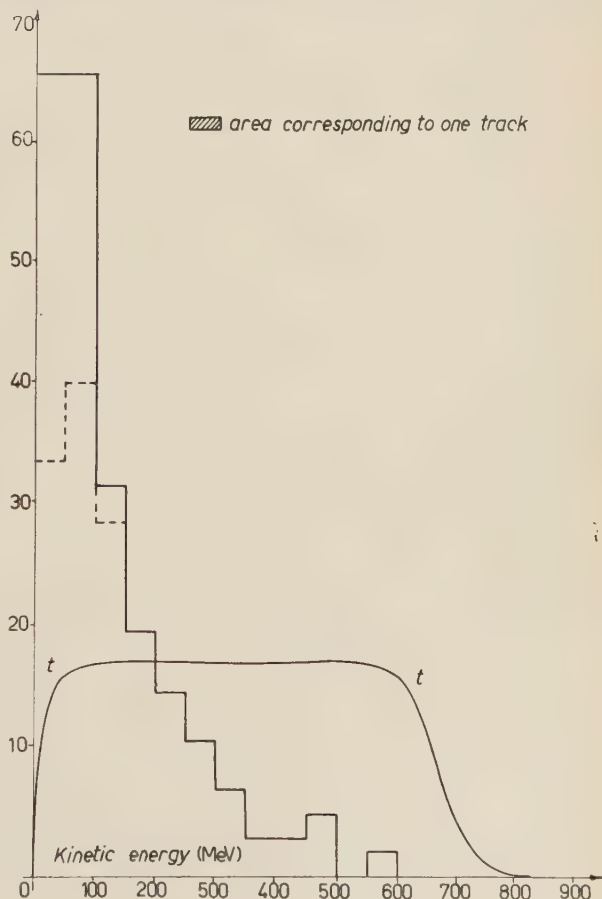


Fig. 2. - Energy distribution of the charged Σ hyperons in the laboratory system.

$K^- + N \rightarrow \Sigma + \pi$, and that their angular distribution is isotropic in the centre of mass system. This t curve takes into account the fact that the collided nucleon is bound in a nucleus, and it has been normalized in such a way that it refers to the same number of particles as the full line hystogram.

One notes immediately the big number of Σ -hyperons emitted with low energy. As we already said in Section 3, all the tracks that can be due to Σ -hyperons have been followed and carefully analysed, so that we can exclude that a loss of fast hyperons has occurred.

The observed shape of the energy spectrum can be due to three facts:

a) Inelastic scattering of the charged Σ -hyperons inside the nucleus. — As discussed in the following (Section 7), we can give an estimate of the probability of Σ inelastic re-emission, and we see that re-emissions can occur in about $12\% \div 18\%$ of all Σ interactions. We know from the works on K^- at rest ⁽¹⁵⁾ and on K^- of low energy ⁽⁶⁻⁸⁾ that about 60% of the produced Σ -hyperons are absorbed in the nucleus in the reaction: $K + N \rightarrow \Lambda^0 + N$. From the above figures we derive that $8\% \div 13\%$ of the emitted Σ -hyperons have suffered an inelastic scattering inside the nucleus.

b) Emission of more than one π -meson. — As will be discussed in Section 6, the emission of two π -mesons occurs, as a maximum, in $13\% \div 14\%$ of the stars. This percentage, at the present point of our research, is not very well established, but it seems approximate enough to say that the effect of double pion emission is not sufficient to explain the observed feature of the Σ -hyperon energy spectrum.

c) Backward emission in the centre of mass system. — To check this last assumption, we have to calculate the angular distribution in the centre of mass system. The uncertainties in this calculation derive from the momentum distribution of the nucleons in the nucleus and from the possibility of a not negligible occurrence of reactions with two or more nucleons. The indications we have so far on this point are very poor: it seems that this type of reaction is very rare in the interactions of K^- -mesons in flight ⁽⁶⁾, while it probably occurs in 20% of the cases in the capture of K^- -mesons at rest ⁽⁵⁾.

Nevertheless the calculation of the angular distribution of the Σ -hyperons in the centre of mass system has been carried out, under the assumption that the production reaction occurs with a single nucleon, and for three different β values of the centre of mass ($\beta_c = 0.525$, corresponding to the reaction with a free proton; $\beta_c = 0.618$ and $\beta_c = 0.430$, corresponding to the reactions with a bound nucleon having, respectively, a Fermi momentum in the opposite direction or in the same direction of the incoming K -meson). The results are given in Fig. 3, and indicate that the angular distribution is strongly peaked backward, even in the more unfavorable case of $\beta_c = 0.430$.

The effect of inelastic scattering of the Σ -hyperons inside the nucleus, that

could produce the observed asymmetry, has been found to be small (see point *a*) of this section), while the elastic scatterings, if present in non negligible percentage, would have only the effect of destroying any asymmetry.

If the percentage of reactions with two nucleons is appreciable, the angular distribution calculated in the described way would be wrong as, in this case, the β values of the centre of mass are different from the ones previously used. (They become, respectively: $\beta_c = 0.369$; $\beta_c = 0.503$ and $\beta_c = 0.232$, if we make the calculation in a very rough way, *i.e.*, if we assume that the K^- -meson collides with a particle of mass equal to two nucleon masses. In this approximation, one takes into account only the relative motion of the two nucleons, considered as a whole, with respect to the K^- -meson, neglecting the relative motion of the two nucleons one with respect to the other.) One way of selecting the possible cases of reactions with two nucleons is to look after those events for which the momentum of the Σ -hyperon in the

centre of mass system, calculated under the assumption of one nucleon interaction, is greater than the maximum value allowed by the available energy. We found 18 of these cases out of the 170 Σ -hyperons observed. Most probably this percentage is not high enough to affect seriously the calculated angular distribution; nevertheless we calculated the angle of emission in the centre of mass system of these 18 Σ -hyperons using the β values corresponding to the reaction with two nucleons and we found that 17 of them are emitted backward.

We take as established that the Σ -hyperons that we observe are prevalently emitted backward in the centre of mass system. We don't know if

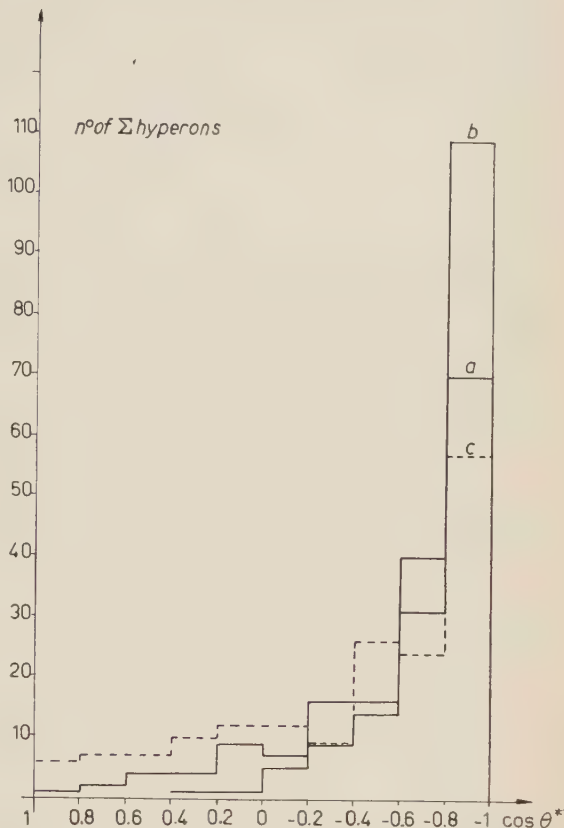


Fig. 3. - Angular distribution of the charged Σ hyperons in the centre of mass system: *hystograms* *a*, *b*, *c*, correspond, respectively, to $\beta_c = 0.525$, $\beta_c = 0.618$ and $\beta_c = 0.430$.

this effect is a feature of the production mechanism in the elementary reactions listed in the Introduction, or an effect of the subsequent mechanism of emission from the nucleus. In the first case, a backward distribution must be observed also in similar experiments with hydrogen bubble chamber; the data at present available to us ⁽⁹⁾ are not concerned with this problem. In the second case, one must think that the production in a nucleus takes place in such a way that the Σ -hyperons emitted forward have a longer path in nuclear matter, and consequently a greater probability to be absorbed.

6. - Analysis of the π -mesons emitted from the parent stars of Σ -hyperons.

All the prongs of the stars emitting Σ -hyperons have been followed; the ones that do not come to rest in the stack have been studied with ionization and scattering measurements.

The results concerning the emission of charged π -mesons together with a charged hyperon are collected in the following Table.

n ^o . of charged pions hyperons										
	0	π^+	π^-	π^\pm	$\pi^+\pi^-$	$\pi^-\pi^-$	$\pi^+\pi^+$	$\pi^\pm\pi^\pm$	$\pi^-\pi^\pm$	$\pi^+\pi^\pm$
Σ^-	26	2	0	12	0	0	0	1	0	0
Σ^+	18	0	2	9	0	1	0	1	0	0
Σ^\pm	49	0	7	28	2	0	0	3	4	2

These data refer to 167 parent stars of Σ -hyperons; three cases of observed Σ -hyperons have not been included, owing to the fact that they are probably cases of production of three strange particle (see Appendix III).

One can see that 14 out of 167 events show a double production of charged π -mesons; the percentage that one can derive ($14/167 = 8.4\%$) is certainly an underestimation, as one does not take into account the probability of absorption inside the nucleus and the π^0 production.

An evaluation of the production of π^0 -mesons can be given looking at the features of all the studied stars, because in this case, as we take into account all the reactions produced by the incoming K^- -mesons, we can say that the ratio π^0/π^\pm has the value $\frac{1}{2}$.

We hope to be able to give a better estimate in a second paper; now, in order to derive the total number of π^\pm emitted from the analysed stars, we must take as good enough the evaluation of about 350 cases of K^- re-emissions given in Section 8. From the data given in Sections 3 and 8, one can easily derive that the total number of π^\pm -mesons emitted is 552. Taking

50 % of absorption probability ($6 \cdot 12$) for the charged π -mesons and the value $\frac{1}{2}$ for the ratio $(\pi_{\text{produced}}^0)/(\pi_{\text{produced}}^\pm)$, we obtain that the total number of neutral and charged π -mesons produced in the 1675 studied stars is 1656.

From the above figures one can see that, even in the extreme hypothesis that the K re-emissions (with or without charge exchange) occur without π -meson production, the percentage of hyperons emitted together with two pions cannot be much higher than $13 \% \div 14 \%$.

7. - Interactions of charged Σ -hyperons.

The total track length of the 170 recognized Σ -hyperons is 225 cm. Two Σ -hyperons have been observed to interact in flight showing the re-emission of a charged Σ -hyperon. We have certainly missed all the Σ -interactions that were not visibly different from proton interactions.

To evaluate this type of loss, we looked at all the interactions found following the prongs of the studied stars. We divided them in groups with respect to the distance of the secondary interactions from the primary star: taking into account the range distribution of the prongs of our stars, it is easy to calculate which distribution has to be expected for the number of interactions if all the interacting particles were protons. The deviations from the expected distribution are due to the fact that between the interacting particles there are also Σ -hyperons, which contribute only to the interactions found near the primary star, owing to their short mean life.

In this way we obtained that the actual number of Σ -hyperons interactions should be compared between 11 and 17 (the uncertainty is due to the roughness of the calculations).

From the preceding figures we derive that the number of observed Σ -hyperons must be increased by $\sim 10 \%$ to take into account the loss due to non recognized Σ interactions (this correction has been taken into account in Section 4).

Furthermore, as we have observed two cases of Σ -interactions with the re-emission of a charged Σ -hyperon, we can say that re-emissions occur probably in $12 \% \div 18 \%$ of all Σ -interactions. This percentage has been used in Section 5, point a)).

8. - K^- re-emission.

In order to evaluate the number of K^- -mesons re-emitted from the analysed stars, we can make the following considerations.

The K^- -mesons emitted with $\beta \leq 0.70$, that come to rest, or interact and

(¹²) M. BLAU and M. CAULTON: *Phys. Rev.*, **96**, 150 (1954).

have been recognized by scattering and ionization measurements, are 33. This number, corrected for the loss of K^- -mesons⁽¹³⁾, becomes 38.

The tracks of the particles emitted with $\beta > 0.70$ (as it has been pointed out in Section 3) have been followed only if they make an angle $\leq 45^\circ$ with the primary direction. The number of tracks selected according to this criterion is 215, and the total track length that has been followed is 1920 cm.

A total of 75 secondary interactions has been found, and 5 of these have been certainly recognized as K^- interactions producing Σ^\pm hyperons.

In the energy interval considered, the cross-section for interactions of π -mesons and of K^- -mesons are nearly equal^(12,14-17); in Section 4 we have seen that the probability for the K^- -mesons to produce a star by which a recognized Σ -hyperon is emitted, is $\sim 10\%$; we can then estimate that 50 of the secondary interactions observed are due to K^- -mesons and the remaining 25 to π -mesons. This means that, in the considered cone of 45° around the primary direction, there are, roughly, 143 K^- mesons re-emitted with $\beta > 0.70$.

From this datum we can

derive, approximately, the total number of K^- re-emissions. We plot the number of K^- -mesons, ΔN , that are in a certain interval of energy loss, $\Delta(\Delta E/E)$ (E is the kinetic energy of the incoming K^- -meson) versus the energy loss $\Delta E/E$, and assume that $\Delta N/(\Delta(\Delta E/E))$ is a constant.

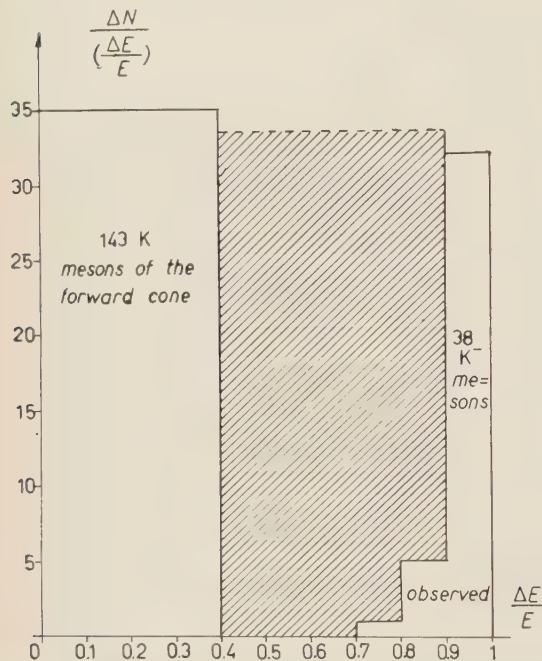


Fig. 4. Number of re-emitted K^- -mesons versus their energy loss.

⁽¹³⁾ G. L. BACCHELLA, A. BERTHELOT, A. BONETTI, O. GOUSSU, F. LEVY, M. RENÉ, D. REVEL, J. SACTON, L. SCARSI, G. TAGLIAFERRI and G. VANDERMAEGE: *Nuovo Cimento*, **8**, 215 (1958).

⁽¹⁴⁾ A. H. MORRISH: *Phil. Mag.*, **45**, 47 (1954).

⁽¹⁵⁾ S. J. LINDENBAUM and L. C. L. YUAN: *Phys. Rev.*, **92**, 1578 (1953).

⁽¹⁶⁾ E. LOHRMANN, M. NIKOLIĆ, M. SCHNEEBERGER, P. WALOSCHEK and H. WINZELER: *Nuovo Cimento*, **7**, 163 (1958).

⁽¹⁷⁾ Present work: Appendix I.

The plot is shown in Fig. 4. The dashed part represents the number of K^- -mesons that we do not recognize; it is about 160. The total number of K^- re-emitted results to be ~ 350 .

This is only a first, rough evaluation of the K^- re-emissions. On this subject, further experimental work is in progress in our laboratory. We hope to be able to give, in a second paper, the results of the following through and of the scattering measurements of nearly all the tracks of particles emitted with $\beta > 0.70$ so that a separation between the π -mesons and the K -mesons can be made on a more reliable way.

9. - Conclusions.

As we already said in the Introduction, in this work we are mainly concerned with the problem of charged Σ -hyperons emission from 1.15 GeV/c K^- interactions.

We have studied 1675 stars; correcting for 10 % of incoming π^- -mesons the studied K^- interactions result to be 1510. We summarize here the most important results obtained.

$$\text{Percentage of } \Sigma^- \text{ hyperons emitted } \frac{186}{1510} = 12,3\%$$

$$\text{Percentage of } \Sigma^+ \text{ hyperons emitted } \frac{64}{1510} = 4,1\%$$

$$\text{Ratio } \frac{\Sigma_{\text{emitted}}^-}{\Sigma_{\text{emitted}}^+} = 2,9$$

$$\text{Ratio } \frac{\Sigma_{\text{re-emitted}}}{\text{all } \Sigma_{\text{interactions}}} \approx 0,12 \div 0,18$$

Moreover we have seen that, in the laboratory system, the energy distribution of the charged Σ -hyperons is peaked toward the low energies. It seems probable, as discussed in Section 5, that this fact is due to a backward emission of the charged Σ -hyperons in the centre of mass system.

We evaluate also, very roughly, the percentage of K^- re-emissions: $\sim 350/1510 \approx 23\%$; this makes possible to give, as an order of magnitude, the percentage of neutral strange particles (σ^0 , Λ^0 , Σ^0) emitted from the studied stars: $\sim 60\%$.

Data on the production processes have not yet been derived, both for the fact that not all the experimental results that one can obtain from the material at our disposal have been collected, and for the fact that no detailed results on 1.15 GeV/c K^- interactions on hydrogen are available to us.

The purpose of the work that is in progress in our laboratory is to complete measurements on the energy spectra of the emitted π -mesons and K^- -mesons, and to evaluate the percentage of the emitted neutral hyperons.

* * *

The exposure has been carried out at the Berkeley Bevatron; our thanks are due to Prof. E. J. LOFGREN, Prof. E. SEGRÈ, Prof. G. GOLDBERGER and Prof. S. GOLDBERGER.

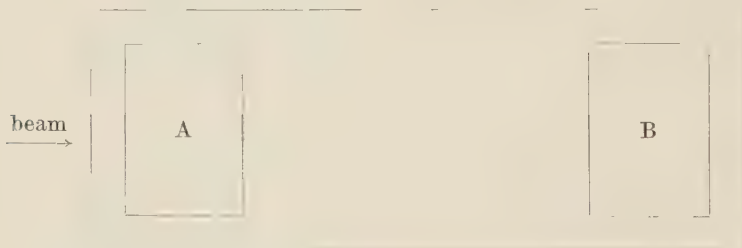
We are also indebted to Dr. L. TALLONE for her work in Berkeley during the exposure and for many useful discussions.

We are grateful to the scanning team of this laboratory, and especially to Mr. G. ALGOSTINO, Mr. N. BORRELLI, Mrs. B. FACCHIN, Mr. M. GRECO and Mr. P. TROSSERO, for their accurate work in the following of the tracks and valuable help in the measurements.

APPENDIX I.

The region A ($3\text{ cm} \times 5\text{ cm}$; top of the stack) has been explored for stars in 5 sheets; the region B ($3\text{ cm} \times 5\text{ cm}$; bottom of the stack) in 7 sheets.

Moreover the tracks of the beam that enter region A and region B have been counted.



The numbers of stars per cm^3 of emulsion are:

$$92 \pm 2 \text{ stars/cm}^3 \quad \text{in the region A}$$

$$14 \pm 2 \text{ stars/cm}^3 \quad \text{in the region B}$$

The flux values are:

$$0.88 \times 10^4 \text{ tracks/cm}^2 \quad \text{entering region A}$$

$$0.265 \times 10^4 \text{ tracks/cm}^2 \quad \text{entering region B.}$$

The mean distance between regions A and B is 19.5 cm.

If we call σ_A and σ_B the apparent cross-sections in the regions A and B, x_A and x_B the percentages of the $(K+\pi)$ component in the regions A and B,

and we assume $\sigma_\mu = 0$, we have:

$$\begin{aligned} \sigma_A &= x_A \sigma_{K+\pi} & \frac{\sigma_A}{\sigma_B} &= \exp \left[\frac{19.5}{\lambda_{K+\pi}} \right] = 1.95_{-0.27}^{+0.35} . \\ \sigma_B &= x_B \sigma_{K+\pi} \end{aligned}$$

From the preceding expressions one derives:

$$\lambda_{K+\pi} = 29_{-6}^{+9} \text{ cm} ; \quad x_A = 0.30_{-0.07}^{+0.1} .$$

APPENDIX II

a) Correction of the number of observed Σ^- captured at rest.

The evaluation of the correction to be applied to the number of observed Σ^- captured at rest is based on the data given by the Bern group ⁽⁶⁾ and by the K-collaboration group ⁽⁵⁾. They studied the prongs distribution of 79 certain Σ^- absorptions at rest, and obtained the following results:

	No. of Σ^- captures with zero prongs	No. of Σ^- captures with one prong	Total no. of Σ^- captures
Bern group ⁽⁶⁾	7	2	16
K-collaboration ⁽⁵⁾	36	12	63

Taking a mean value of the above figures, we found that 72 % of all Σ^- captures are captures with zero or one prong. It results that the corrected number of Σ^- hyperons emitted from the stars studied in the present work, and captured at rest, is 75.

b) Correction of the number of observed $\Sigma^+ \rightarrow p$ decays.

Of the 18 $\Sigma^+ \rightarrow p$ decays observed, 13 have the decay proton with a β value higher than the β value of the decaying hyperon. We start from the assumption that we do not lose any event of this type, and we try to evaluate what is the corresponding number of cases that are expected to have the decay proton with a β value lower than the β value of the decaying hyperon. This number (if we assume an isotropic distribution of the decay protons in the centre of mass system) depends only on the β value of the Σ -hyperon at its decay. The mean value of β_{decay} , (calculated on 135 cases of Σ -hyperons observed to decay in flight, is 0.40. From this figure we deduce that the observed 13 cases of $\Sigma^+ \rightarrow p$ decays, having the decay proton with β value higher than the β value of the decaying hyperon, represent 56 % of all the $\Sigma^+ \rightarrow p$ decays in flight. It follows that the total number of $\Sigma^+ \rightarrow p$ decays in flight that we should observe is 23; to this number we must add the 6 cases of observed $\Sigma^+ \rightarrow p$ decays at rest.

c) **Attribution of the sign of the electric charge to the decaying Σ -hyperons.**

We have calculated that we should have observed 29 cases of $\Sigma^+ \rightarrow p$ decays; we assume that we must have an equal number of $\Sigma^+ \rightarrow \pi^+$ decays. This means that to 21 of the 94 cases of decays with undetermined charge must be attributed a positive charge, and to the remaining 73 a negative charge.

APPENDIX III

a) **Interactions of Σ -hyperons.**

We describe here the two cases of Σ interactions with re-emission of the Σ -hyperon, that have been observed during the present research.

Event r 1

visible prongs of the parent star:	$\left\{ \begin{array}{l} 5 \text{ stable prongs} \\ 1 \pi^\pm \text{ of kinetic energy } 130 \text{ MeV} \\ 1 \Sigma^+ \end{array} \right.$
distance of Σ^\pm interaction from the parent star:	4.3 mm
mode of decay of the Σ^+ re-emitted:	$\Sigma^+ \rightarrow \pi^+ + n$
visible prongs of the Σ^\pm interaction:	$\left\{ \begin{array}{l} 1 \text{ evaporation track} \\ 1 \Sigma^+ \text{ re-emitted} \end{array} \right.$
kinetic energy of the interacting Σ^\pm :	272 MeV
kinetic energy of the re-emitted Σ^+ :	202 MeV
percentage of energy loss of the Σ :	26 %

Event r 2

visible prongs of the parent star:	$\left\{ \begin{array}{l} 3 \text{ stable prongs} \\ 1 \pi^- \text{ of kinetic energy } 90 \text{ MeV} \\ 1 \Sigma^\pm \end{array} \right.$
distance of Σ^\pm interaction from the parent star:	22 mm
mode of decay of the Σ^+ re-emitted:	$\Sigma^+ \rightarrow p + \pi^0$ (*)
visible prongs of the Σ^\pm interaction:	$\left\{ \begin{array}{l} 2 \text{ evaporation tracks} \\ 1 \text{ proton of kinetic energy } 67 \text{ MeV} \\ 1 \Sigma^+ \text{ re-emitted} \end{array} \right.$
kinetic energy of the interacting Σ^\pm :	490 MeV
kinetic energy of the re-emitted Σ^+ :	135 MeV
percentage of energy loss of the Σ :	72 %

(*) The π^0 meson decays according to the scheme: $\pi^0 \rightarrow e^+ + e^- + \gamma$.

We wish to point out that most probably the two described cases of Σ^\pm interactions with Σ^+ re-emission are due to positively charged Σ hyperons.

b) Production of three strange particles.

We do not observe any certain case of reactions of the type: $K^- + \text{nucleus} \rightarrow K^0$ (or K^+) + 2 particles of strangeness -1 .

Two events that could probably be interpreted in this way are described in the following.

Event t 1: A K^- of the beam interacts giving rise to a star with the following visible prongs:

3 stable, prongs, of range respectively, 150 μm , 300 μm , 18 mm;

1 Σ^- hyperon, that comes to rest after 10.2 mm and gives rise to a capture star with two prongs;

1 Σ^+ hyperon, that decays in the mode $\Sigma^+ \rightarrow p + \pi^0$ after 19.4 mm.

In this case, the uncertainty comes from the fact that the event interpreted as the decay $\Sigma^+ \rightarrow p + \pi^0$ could be the scattering of a proton.

Event t 2: A K^- of the beam interacts giving rise to a star with the following visible prongs:

5 stable prongs, of range, respectively, 200 μm , 350 μm , 800 μm , 800 μm , 4 mm;

1 Σ^\pm hyperon, that decays in the mode $\Sigma^\pm \rightarrow \pi^\pm + n$ after 9.9 mm;

1 HF (or Σ^- or K^-) that gives rise to three prongs of range, respectively, 100 μm , 1.2 mm and 9 mm, after a range of 1 μm .

The uncertainty, in this case, comes from the fact that, due to the very short range, it is not possible to exclude that the HF (or Σ^- or K^-) is a π^- -meson.

RIASSUNTO

Lo scopo del presente lavoro è di dare alcuni risultati ottenuti dall'analisi di 1675 interazioni di mesoni K^- di alta energia con i nuclei dell'emulsione. I principali risultati riguardano l'emissione di iperoni carichi, la loro distribuzione energetica ed angolare, il rapporto $(\Sigma^- \text{ emesse})/(\Sigma^+ \text{ emesse})$; viene data anche una valutazione della probabilità di rimissioni inelastiche delle Σ .

LETTERE ALLA REDAZIONE

(La responsabilità scientifica degli scritti inseriti in questa rubrica è completamente lasciata dalla Direzione del periodico ai singoli autori)

Remarks on Compound Models, Conserved Currents and Weak Interactions.

M. E. MAYER

Physico-Mathematical Faculty, Parhon University - Bucharest

(ricevuto il 12 Luglio 1960)

During the past few years there has been revived interest in compound models for elementary particles, or at least in determining the minimal number of irreducible fields, that are able to reproduce the known symmetries of known particles⁽¹⁾.

On the other hand, the success of the Feynman, Gell-Mann, Marshak and Sudarshan theory of weak interactions has again raised the question whether weak interactions cannot be consistently interpreted by means of a heavy vector meson, coupled to the conserved V and A currents of the fermions⁽²⁾, and to what extent the four fermion interaction is valid at high energies⁽³⁾.

We wish to discuss briefly some implications of the new symmetry principle, conjectured by GAMBA, MARSHAK

and OKUBO⁽⁴⁾, in connection with the above mentioned problems. GAMBA, MARSHAK and OKUBO (= GMO) have observed that, at least as far as the weak interactions are concerned the three baryons Λ^0, n, p are equivalent to the three leptons μ^-, e^-, ν , in the sense that any reaction permitted or observed for one of the two groups is permitted for the second and conversely, no reaction forbidden for one of the groups has been observed in the other. This permitted the extension of the notions of isospin and strangeness to leptons and led to the following expression of the electric charge in terms of the projection of isospin, T_3 and the baryon and lepton numbers B and L :

$$Q = T_3 + (S + B - L)/2.$$

The electron and neutrino form an isodoublet with $S=0$ and the muon is an isosinglet with $S=-1$, as the Λ^0 .

What is remarkable, is that the three baryons entering the GMO-principle are the fundamental particles of the Sakata-Okun model. Now, the baryon and lepton gauge groups each

⁽¹⁾ S. SAKATA: *Prog. Theor. Phys.*, **16**, 686 (1956); L. B. OKUN: *Proc. Intern. Conf. on High Energy Phys. at CERN*, p. 223 (Geneve, 1958); F. GÜRSEY: *UCRL* 8290; W. THIRRING: *Nucl. Phys.*, **10**, 97 (1959) and *CERN Preprint* (1959).

⁽²⁾ J. SCHWINGER: *Ann. Phys.*, **2**, 407 (1957); M. E. MAYER: *Physik. Verhandl.*, **9**, 57 (1958); A. SALAM and J. C. WARD: *Nuovo Cimento*, **11**, 568 (1959); S. GLASHOW: *Nucl. Phys.*, **10**, 107 (1959).

⁽³⁾ D. I. BLOHINČEV: *Nuovo Cimento*, **9**, 925 (1958); B. L. IOFFE: *Žurn. Eksp. Teor. Fiz.*, **38**, (1960).

⁽⁴⁾ A. GAMBA, R. MARSHAK and S. OKUBO: *Proc. Nat. Acad. Sci. USA*, **45**, 881 (1959).

lead to a conserved current and to a vector field coupled to the respective particles⁽⁵⁾. It is tempting to assume that the two vector mesons introduced so are identical (this would extend the GMO principle also to the symmetry properties of baryons and leptons: the baryons and leptons gauge transformations are the «GMO-transform» of each other) and that weak interactions are mediated by these particles. By complicating the model a little, one could probably obtain the strong interactions between the particles of the Sakata-Okun model in the same manner, provided the coupling constant to baryons is much larger than the one to leptons, or for strangeness-nonconserving processes. This would lead to a six-field model of elementary particles, with $\nu, e, \mu, p, n, \Lambda$ as the fundamental ones while all others are compound ones. The question remaining open, until one can handle such models quantitatively, is: why only three of these particles are stable?

Another possible model (at least as far as charge properties are involved) can be built up out of three fermions and one boson. We may take as fundamental the three leptons ν, e^-, μ^- and a boson field B^+ , carrying electric, baryonic and (negative) leptonic charges. For simplicity assume B scalar, although we do not investigate here spin and parity problems. The baryons are compound particles, their mass being due

mainly to the mass of B (note that the mass differences between the three leptons and the three baryons entering the GMO-symmetry are of the same order of magnitude). One thus obtains the following scheme of particles:

$$\begin{aligned} p &= (\nu B^+), & n &= (e^- B^+), \\ \Lambda &= (\mu^- B^+), & \pi^+ &= (\nu B^+ B^- e^+), \\ \pi^- &= (\bar{\nu} B^- B^+ e^-), \\ K^+ &= (\nu B^+ B^- \mu^+), \\ \Sigma^+ &= (\nu B^+ B^- e^+ B^+ \mu^-), \\ \Xi^- &= (\nu B^- B^+ \mu^- B^- \mu^-), \text{ etc.} \end{aligned}$$

As regards the «high-energy limit» of weak interactions, *i.e.* the energy at which weak interactions become comparable to electromagnetic ones, or weak radiative corrections become comparable with the effects (*cf.* ref.⁽³⁾ and unpublished calculations by the author, quoted there) this problem becomes senseless in a compound model, or in a model with intermediary bosons. However it must again be stressed, that both the four-fermion interaction and the intermediary boson descriptions are rather naive manners of expressing the observed low-energy behaviour of weak interactions. The same applies, *mutatis mutandis*, to various «compound» models, which certainly express no more than some symmetry principles. What is needed urgently is a method to handle quantitatively the so-called bound and unstable states.

A detailed account of the questions touched upon here is in preparation.

⁽⁵⁾ See for example, M. E. MAYER: *Nuovo Cimento*, **11**, 760 (1959).

Ξ^- Production in Emulsion by 1.1 GeV/c K^- Mesons.

M. BALDO CEOLIN, A. CAFORIO (*), O. FABBRI, F. FARINI,
A. FERILLI (*), G. MIARI and J. SCHNEPS (**)

Istituto di Fisica dell'Università - Bari

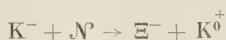
Istituto di Fisica dell'Università - Padova

Istituto Nazionale di Fisica Nucleare - Sezione di Padova

(ricevuto il 18 Luglio 1960)

A stack of G-5 emulsions ($(4.2 \times 20 \times 15) \text{ cm}^3$) has been exposed to K^- -mesons of 1.1 GeV/c momentum. The separated K^- beam from the Berkeley bevatron was designed and built by GOOD and TICHON for the Alvarez Hydrogen bubble chamber. The stack was exposed behind this chamber during its operation.

The research has been undertaken to verify the reaction



and to study the frequency of production of Ξ^- particles.

A total of 225 stars have been found by scanning along the beam tracks. These interactions have been analyzed completely for the study of the K^- -nucleon interaction. This work has been completed and the results will be presented in a forthcoming paper.

In addition 1030 stars have been found by « area scanning », of which the secondary particles with grain density greater than 1.5 times the « plateau value » and dip angles $< 30^\circ$ were analysed. It is to be emphasized that the Ξ^- hyperons will in general have relatively high velocities and corresponding small angles in the laboratory system, because the incoming K^- energy is near threshold. The criteria for angles and ionization were chosen basing on these considerations.

From the analysis of the 1255 stars, 45 tracks have been found, which decayed in flight into π -mesons.

Measurements of both ionization and $p\beta$ on the track of the decaying particle and on the secondary particle were made when possible. In a few cases measurements of the range were also possible.

For 28 events on which it has been possible to make combined measurements on both the primary and the secondary particles, we have discriminated the decay of 27 Σ hyperons and 1 Ξ^- particle from kinematic considerations.

For 8 events we could measure the ionization of the primary particle, but

(*) A. CAFORIO and A. FERILLI: Institute of Physics, University of Bari, wish to acknowledge the C.N.R. for a research grant.

(**) On leave from Tufts University, Medford, Mass.

not the $p\beta$; for these events it was possible to make good measurements on the secondaries and to identify the decaying particles as Σ from kinematics.

For the 9 other events, on the track of the secondary particle it was possible to make measurements only of the ionization at the point of decay and near the point where it left the stack. Knowing this distance we were able to give limits for the momentum of the secondary particle. For 5 of these 9 events, the measurements on the primary of which were satisfactory, we have recognized from

The data concerning the two decays $\Xi^- \rightarrow \Lambda^0 + \pi^-$ are given in Table I.

The Q -value of the event 117 T is found to be:

$$Q = 65 \pm 8 \text{ MeV}.$$

In the event 392 A the range of the primary is particularly short, while the secondary stops in emulsion and is a π^- .

From the momentum of the π and the angle of the decay the ionization of the primary has been calculated on our calibration curves assuming that it is a

TABLE I.

Star	Type	Visible energy of outgoing particles in MeV	Decaying particle			
			Range in cm	Dip	Angle between incoming K^- and Ξ	Momentum of Ξ at production
117 T	0+3K	247 ± 33	2.6	13°	14°	840 ± 60
392 A	0+3K	94_{-2}^{+17}	0.041	29°	42°	414 ± 56
			Secondary particle from decay			
Momentum at decay in MeV/c	Time of flight in 10^{-11} s	Angle between Ξ and π	Dip	Momentum at decay point	Length followed mm	
770 ± 70	$14.22_{-0.87}^{+0.99}$	41°	24°	220 ± 20	31 out	
414 ± 56	$0.44_{-0.05}^{+0.07}$	119°	55°	100 ± 5	17.5 stop	

kinematic considerations 3 Σ and 2 K decays. 3 of the 4 remaining events fit with greater probability the decay $\Sigma^\pm \rightarrow \pi^\pm + n$ and the last event 329A is probably a Ξ^- decay.

By the analysis we conclude that 41 decays were $\Sigma^\pm \rightarrow \pi^\pm + n$, 2 were $K^- \rightarrow \pi^- + \pi^0$, 1 was $\Xi^- \rightarrow \Lambda^0 + \pi^-$ (event 117 T) and in addition one was very probably $\Xi^- \rightarrow \Lambda^0 + \pi^-$ (event 329 A). No event has been found which fits the decay $\Xi^- \rightarrow n + \pi^-$.

Σ^- , or a Ξ^- or a $K^- \rightarrow \pi^- + \pi^0$. The values found are the following:

$$I_{\Xi} = 55_{-7}^{+8} \text{ grains/50 } \mu\text{m}$$

$$I_{\Sigma} = 25_{-3}^{+2} \text{ grains/50 } \mu\text{m}$$

$$I_K = 23.5 \pm 1.5 \text{ grains/50 } \mu\text{m}$$

which are to be compared with the measured value of the ionization:

$$I = 52 \pm 5 \text{ grains/50 } \mu\text{m}.$$

Even if there are large fluctuations, the identification of the event as the decay of a hyperon Ξ is very probable

We cannot completely exclude the decay $K^- \rightarrow \pi^- + \pi^0 + \pi^0$ although it seems very unlikely; in fact, we obtain for the momentum of the π in C.M.S., assuming that the decaying particle is a K-meson

$$p = 133 \pm 9 \text{ MeV}/c$$

while the maximum momentum for the decay $K^- \rightarrow \pi^- + \pi^0 + \pi^0$ is

$$p = 133 \text{ MeV}/c.$$

As for the frequency of Ξ^- production in the reaction K-nucleus, it should be mentioned that in the study of the stars there has been found one event which is interpretable as the emission of a cascade hyperfragment. The event appears as a triple-centered star; the interpretation of this event is unfortunately not secure since the prongs are a few microns long ($\sim 5 \mu\text{m}$) and the identification of their nature was not possible.

Since the value of the lifetime of the mixture $\Sigma^\pm \rightarrow \pi^\pm + n$ reported in the lite-

rature from measurements in emulsion ⁽¹⁾, seems to present some anomalies, we have calculated the lifetime of our sample with the method of BARTLETT ⁽²⁾.

The value we have found with a confidence limit corresponding to a probability of 95%, is:

$$\tau = 0.73^{+0.40}_{-0.19} \cdot 10^{-10} \text{ s}.$$

In the analysis of our stars we have found also 10 tracks of baryonic mass which disappear in flight. If we add them to the above 41 decays of the type $\Sigma^\pm \rightarrow \pi^\pm + n$, we obtain:

$$\tau = 0.80^{+0.34}_{-0.20} \cdot 10^{-10} \text{ s}.$$

* * *

The authors wish to thank those who helped to make the experiment possible by exposing the stack, namely Professors M. GOOD, H. TICHO, U. CAMERINI, W. F. FREY and D. PROWSE.

⁽¹⁾ See for example the data reported in: *CERN Conference on High Nuclear Physics* (Geneva, 1958) and K^- COLLABORATION: *Nuovo Cimento*, **15**, 873 (1960).

⁽²⁾ M. S. BARTLETT: *Phil. Mag.*, **44**, 249 (1953).

Prong Analysis of Multiple Production of Pions at Bevatron Energy According to a Statistical Theory.

F. CERULUS and J. VON BEHR

CERN - Geneva

(*Nuovo Cimento*, **16**, 1046 (1960))

ADDENDA

The captions of the individual figures have unfortunately been omitted in the publication. They read as follows:

- Fig. 1. - p-spectrum of 2 prong events.
- Fig. 2. - π^\pm -spectrum of 2 prong events.
- Fig. 3. - $p + \pi^\pm$ -spectrum of 2 prong events.
- Fig. 4. - p-spectrum of 4 prong events.
- Fig. 5. - π^\pm -spectrum of 4 prong events.
- Fig. 6. - $p + \pi^\pm$ -spectrum of 4 prong events.
- Fig. 7. - p-spectrum of 6 prong events.
- Fig. 8. - π^\pm -spectrum of 6 prong events.
- Fig. 9. - $p + \pi^\pm$ -spectrum of 6 prong events.
- Fig. 10. - p-spectrum of all events.
- Fig. 11. - π^\pm -spectrum of all events.
- Fig. 12. - $\pi^\pm + p$ -spectrum of all events.

LIBRI RICEVUTI E RECENSIONI

L. EISENBUD and E. P. WIGNER -
Nuclear Structure. Princeton, University Press, 1958, pp. 128.

Lo scopo di questa breve monografia è di introdurre il lettore alle principali idee con cui oggi vengono affrontati i moltissimi problemi particolari offerti dalla fisica nucleare. Se si pensa alla mancanza di sistematicità e al rapido sviluppo che sono propri della ricerca in questo campo, un'impresa del genere appare molto difficile. Il risultato ottenuto dai due autori, il cui nome non richiede alcuna illustrazione, ci sembra veramente brillante: l'esposizione delle idee fisiche più interessanti delle diverse teorie nucleari raggiunge una chiarezza e precisione difficilmente riscontrabile nella letteratura precedente non specializzata. L'uso della matematica è ridotto a poche formule generali e nella esposizione si fa sempre appello piuttosto alla immaginazione del lettore che non alla sua abilità nel maneggiare espressioni complicate. Ciò dovrebbe ren-

dere particolarmente gradito il volume a tutti i non specialisti di fisica nucleare che vogliono in breve tempo essere messi in grado di affrontare opere più dettagliate o più semplicemente essere informati sull'argomento.

Diamo ora un breve schema delle varie parti del libro. Nei primi tre capitoli vengono discusse in modo organico le proprietà più elementari dei nuclei e cioè energie di legame, tipi di disintegrazioni spontanee, spin, momenti angolari e dimensioni dei nuclei nel loro livello fondamentale. Nel quarto capitolo sono descritte le reazioni nucleari possibili. Dopo un esame delle forze tra due nucleoni (capitolo 5), i modelli nucleari finora inventati sono descritti nei capitoli 6, 7, 8. Due capitoli 9, 10 sono dedicati ai modelli utili per lo studio delle reazioni. L'ultima parte del libro è dedicata alla interazione β e alle transizioni elettromagnetiche. Una vasta bibliografia divisa per argomenti è inclusa alla fine del volume.

G. JONA-LASINIO

PROPRIETÀ LETTERARIA RISERVATA

Direttore responsabile: G. POLVANI

Tipografia Compositori - Bologna

Questo Fascicolo è stato licenziato dai torchi il 12-IX-1960